

Quantitative analysis of changes in antioxidant in crosslinked polyethylene (XLPE) cable insulation material exposed to heat and gamma radiation

Shuaishuai Liu¹, Stephen W Veysey², Leonard S Fifield³, Nicola Bowler^{1*}

¹Department of Materials Science and Engineering, Iowa State University, Ames, IA 50011, USA

²Chemical Instrumentation Facility, Iowa State University, Ames, IA 50011, USA

³Pacific Northwest National Laboratory, Richland, WA 99354, USA

nbowler@iastate.edu*

Key words: Pyrolysis gas chromatography-mass spectroscopy, antioxidant, oxidation induction time (OIT), quantitative analysis, thermal and gamma radiation aging, cross-linked polyethylene

Abstract

Quantitative analysis of the antioxidant poly(1,2-dihydro-2,2,4-trimethylquinoline) (pTMQ) was conducted on pristine, thermally-aged, and gamma radiation-aged commercial cross-linked polyethylene-(XLPE-)based cable insulation material aged at temperatures 60, 90, and 115 °C, with gamma radiation exposure dose rates of 0, 120, 300, and 540 Gy/h for 15 days. The quantification of antioxidant was performed using pyrolysis gas chromatography-mass spectrometry (Py-GCMS). Oxidation induction time (OIT) was measured using differential scanning calorimetry (DSC) and correlation was made between the quantified depletion of antioxidant and measured OIT. It was observed that, in the case of isothermal aging, the quantity of antioxidant and OIT decreased with increasing gamma radiation dose. In the case of samples exposed to the same gamma radiation dose, the quantity of antioxidant and OIT were observed to decrease with increasing aging temperature. Depletion in the quantity of antioxidant relative to that in the pristine material ranged from 7 to 93 % for differently aged samples. The measured decline in OIT ranged from 0 to 80 %. Change in the quantity of antioxidant in the material was observed to follow the same trend as the change in OIT when the samples were aged under various conditions, with a correlation coefficient of 0.82. The observations are explained in terms of the reaction between the antioxidant and free radicals created during exposure of the samples to thermal and gamma radiation.

Introduction

As of February 2017, 61 commercial nuclear power plants (NPPs) with 99 nuclear reactors were operating in the United States [1]. Among the 99 reactors, 59 were pressurized water reactors (PWRs) and 40 were boiling water reactors (BWRs) [2]. Operation of a typical PWR requires thousands of kilometers, and a typical BWR requires hundreds of kilometers, of electrical cables [3, 4]. The study of degradation and failure behavior of cable insulation materials is important because the integrity of the cables' polymeric insulation material is critical for safe and reliable operation of the NPP. The insulation materials suffer degradation through exposure to heat, gamma radiation and other environmental stressors throughout their service lifetime.

Polymer-based materials used for cable insulation are composites, consisting of a polymer matrix and various fillers and additives to protect the material against fire and degradation. Antioxidants (AOs) are important additives that protect the polymer from oxidative aging. Crosslinked polyethylene (XLPE) is one of the most widely used polymers for cable insulation in NPPs due to its superior

properties such as good resistance to thermal and gamma radiation and good mechanical properties [5-8]. Enormous research efforts have been made to study the effect of irradiation such as ultraviolet irradiation, electron beam irradiation and gamma irradiation on polyethylene with or without antioxidants or stabilizers in various fields such as wires and cables, biopharmaceuticals and biotechnology [9-14]. Researchers evaluate antioxidants by measurements such as yellowness index of the material under various irradiation conditions because aging causing the generation of carbonyl bonds which causes the material to appear yellowish in color [9]. To the best of our knowledge, however, there has been no study directly quantifying the changes in antioxidant caused by irradiation. For the application of wires and cables in NPPs, any change in the quantity of antioxidant under different aging conditions may be a key indicator of aging and is important for understanding the degradation behavior of these insulation materials. Evaluation of antioxidant depletion may help to determine the remaining service life of the cable.

In this paper, cross-linked XLPE-based cable insulation material was studied. The composition of this material was determined using a combination of analytical techniques: scanning electron microscopy-energy dispersive X-ray spectroscopy (SEM-EDX), Fourier transform infrared spectroscopy (FTIR), thermogravimetric analysis (TGA), Carbon-Hydrogen-Nitrogen (CHN) combustion, and pyrolysis gas chromatography-mass spectroscopy (Py-GCMS). Details of the method by which the material composition was established have been published elsewhere [15, 16] and the results are summarized in Table 1.

The XLPE-based cable insulation material was aged under various conditions, including three temperatures (60, 90, and 115 °C) and four gamma dose rates (0, 120, 300, and 540 Gy/h) for 15 days. This exposure time corresponds to total gamma doses of 0, 45, 110, and 190 kGy, respectively. The quantity of AO in the samples was analyzed following exposure using Py-GCMS.

Measurement of OIT is one of the evaluation methods of oxidative stability and has been used previously as a measure of oxidative degradation, antioxidant depletion, and antioxidant concentration in various types of polyethylene [17-19]. Little research has been conducted, however, to measure antioxidant depletion *directly and quantitatively* during polymer aging and to study the relationship between quantity of antioxidant and measured value of OIT. In this work, OIT was measured using differential scanning calorimetry (DSC) and correlations were made between the changes in OIT and in antioxidant.

Table 1. Composition of pristine XLPE-based cable insulation material studied, and function of the components.

Component	Approximate weight ratio (%)	Function
Cross-linked polyethylene	60	Polymer matrix
Decabromodiphenyl ether (C ₁₂ Br ₁₀ O)	10	Flame retardants
Octabromodiphenyl ether (C ₁₂ H ₂ Br ₈ O)	10	
Sb ₂ O ₃	17	
ZnS		White pigment
Poly(1,2-dihydro-2,2,4-trimethylquinoline)	2	Antioxidant, metal deactivator
1,3,5-Triazine-2,4,6(1H,3H,5H)-trione	trace	Cross-linking booster

Sample Preparation

White XLPE-based insulation material studied in this work was obtained from commercially-available nuclear-grade instrumentation cable consisting of two 16AWG conductors, a laminated aluminum/polyester shield, a drain wire, and a chlorosulphonated polyethylene (CSPE) jacket (RSCC, product code I46-0021), Figure 1. According to the manufacturer, the XLPE insulation was cross-linked by electron-beam radiation. It was removed from the cables by first stripping off the jacket and other external components from the cable assembly, and then pulling out the conductor from the center of the insulation. The wall thickness of the resulting tubular insulation sample material was 0.70 ± 0.05 mm.

To accomplish accelerated aging, samples were suspended by clips on a rack placed in an oven located in the exposure zone of a Co-60 source in the High Exposure Facility (HEF) at Pacific Northwest National Laboratory. In this way, simultaneous thermal and gamma radiation aging of the samples was achieved. The dose rate to each individual sample was controlled by selecting the position of the sample with respect to the radiation source. Overall, 240 different aging conditions were achieved at three aging temperatures (60, 90, and 115 °C), 25 dose rates (ranging from 0 to 540 Gy/h), and five aging durations (5, 10, 15, 20, and 25 days). From this combination of conditions, samples were exposed to doses of gamma radiation in the range from 0 to 320 kGy. Details of the accelerated aging process and sample preparation can be found in a separate paper [20].

From the full set of 240 sample scenarios, 13 were selected for quantitative analysis of antioxidant, presented in this paper. The detailed aging conditions of the 13 selected sample sets are listed in Table 2.

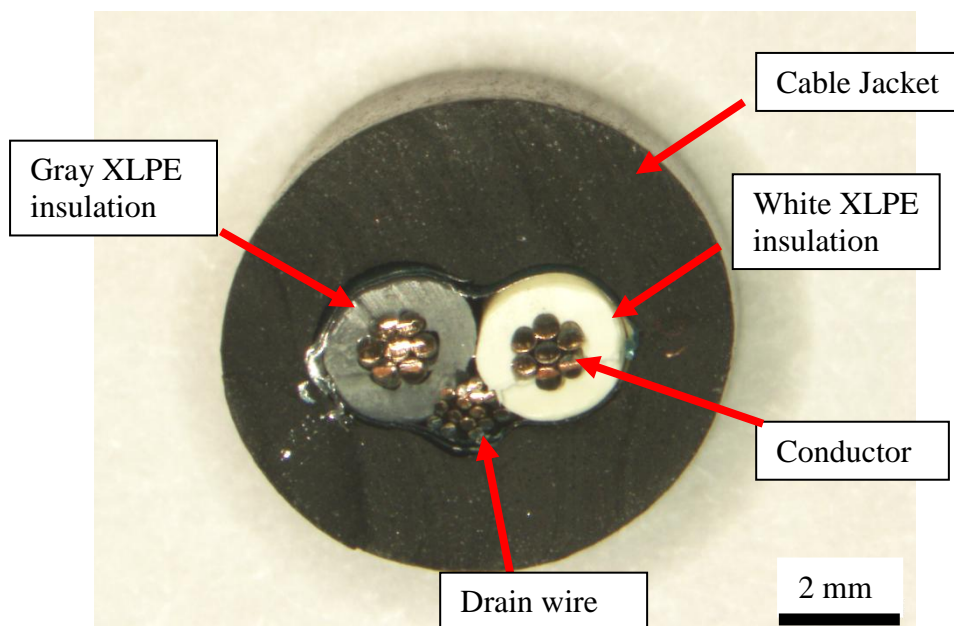


Figure 1. Cross section of the XLPE-based cable insulation material.

Table 2. Aging conditions under which the XLPE-based samples were prepared. Sample ID in the first column is a combination of aging temperature (in °C) and nominal radiation dose (in kGy). All samples other than the pristine sample were exposed for 15 days.

Sample ID	Aging temperature (°C)	Radiation dose rate (Gy/h)	Radiation dose (kGy)
Pristine	Room temp storage	0	0
60C-0	60	0	0
60C-45	60	120	45
60C-110	60	300	110
60C-190	60	540	190
90C-0	90	0	0
90C-45	90	120	45
90C-110	90	300	110
90C-190	90	540	190
115C-0	115	0	0
115C-45	115	120	45
115C-110	115	300	110
115C-190	115	540	190

Materials Characterization Methods

Pyrolysis gas chromatography-mass spectroscopy

The Waters® GCT Premier™ Mass Spectrometer is an accurate-mass time-of-flight (TOF) mass spectrometer coupled to an Agilent 6890 gas chromatograph (GC). The electron ionization (EI) source was used in this study. The Frontier 3030D sample introduction and furnace assembly were mounted on the back inlet of the GC. For thermal desorption studies, the GC column used was a Restex Rxi-5HT, 30 m x 0.25 mm inner diameter x 0.25 micron film thickness. The carrier gas used for all measurements was ultra-high purity helium.

The brominated flame retardants present in the material-under-test, Table 1, are reactive compounds that interact with the surface of deactivated stainless steel sample cups and cause poor measurement reproducibility if stainless steel sample cups are used. In this study, deactivated glass sample cups were used in order to avoid this source of uncertainty. The type ‘Eco-Cup G’ from Frontier Lab was used.

An evolved gas analysis (EGA) test was conducted to determine the evolved temperature profile of each component in the material. The GC column was replaced with a two-meter-long deactivated Silcosteel transfer line for the EGA tests. The GC oven was held at 320 °C. A thin slice of sample (0.5 ± 0.02 mg) was cut from the cross section of the insulation material (Figure 1) and placed into the sample cup. The sample cup was then loaded into the pyrolyzer. The pyrolyzer was held at 80 °C for 2 min, after which the temperature was increased to 800 °C at 20 °C/min. The mass spectrometer total-ion-current (TIC) versus temperature profile of the test material was recorded during the EGA test. This profile was selected with the goal of maximizing signal-to-noise ratio for antioxidant for best quantitative analysis of antioxidant concentration in pristine and differently aged materials. The temperature program targeted complete evolution of the antioxidant from the test sample, while

minimizing contamination to the instrument system from brominated flame retardant and XLPE polymer matrix decomposition residue. The EGA test indicated that holding the sample at 320 °C for 5 min met these objectives. From the TIC versus temperature data, multiple accurate masses were extracted from the TIC data to profile the desorption and pyrolysis of the various components under study.

Thermal desorption (TD) tests were conducted on pristine and differently aged materials using the GC column discussed previously. For accurate quantification of the antioxidant in each sample, 2,5-bis-2-(5-tert-butylbenzoxazolyl)thiophene (BBOT) was used as an internal standard. BBOT was dissolved in acetone at a concentration of 5.5 mg/100 mL. First, 3 μ L of BBOT solution was injected into the sample cup. Then a thin slice of sample (0.260 ± 0.009 mg) was cut from the cross section of the insulation material and added to the sample cup after the acetone had evaporated. The sample cup was then loaded into the pyrolyzer. The pyrolyzer was held at 80 °C for 2 min, then ramped to 320 °C at 20 °C/min. The pyrolyzer was then held at 320 °C for 5 min, according to the EGA test result discussed in the previous paragraph. The GC column was then held at 40 °C for 5 min, followed by a temperature ramp up to 320 °C at 20 °C/min, holding there for 10 min.

The uncertainty of the TD GCMS measurements was estimated by running five replicates on pristine samples with BBOT as the internal standard. The studied compounds were quantified by integrating the corresponding peak areas. Figure 4 shows the weight-normalized AO peak areas of five measurements on pristine and gamma-irradiated sample material. Figure 5 shows the weight-normalized AO peak area on pristine and thermally-aged sample material. The average internal-standard-corrected antioxidant peak area of pristine material is 997, with a standard deviation of 66, which gives an uncertainty of 6.6 %, Table 3. Two replicates were measured for each aged sample scenario. A third replicate was generally measured when the standard deviation of the measured values on the two replicates was more than 6.6 %.

Oxidation Induction Time

Oxidation induction methods are based on the detection of the oxidation exotherm that occurs when a sample is heated in the presence of oxygen. OIT tests were conducted using a TA Instruments Q2000 differential scanning calorimeter. Samples with mass 10.0 ± 0.5 mg were used in each test. The temperature program is shown in Figure 2. The DSC cell was heated at 15 °C/min from 40 °C to 230 °C in nitrogen at a flow rate of 50 ml/min. When the specified temperature of 230 °C was reached, the specimen was held in nitrogen isothermally for 2 min, after which the atmosphere was changed to oxygen maintained at the same flow rate. The specimen was then held isothermally at 230 °C until the oxidative reaction was observed on the thermal curve. The time interval from when the oxygen flow is first initiated to the onset time of the oxidative reaction is referred to as the oxidation induction time. The threshold is defined as 0.1 W/g relative to (above) the baseline and the onset time of the oxidative reaction is defined by the intersection of the test curve with the threshold line, Figure 2. As also can be seen in Figure 2, the melting temperature range of the XLPE sample is from about 90 to 115 °C. The selected temperature for OIT testing, 230 °C, is above the melting temperature range of the tested sample.

Table 3. Internal standard (I.S.) peak area, weight normalized (wt. norm.) antioxidant peak area, and weight normalized antioxidant peak area after correction by the I.S. of five pristine samples.

Replicate	1	2	3	4	5	Mean value	Standard deviation	Uncertainty (%)
I.S. peak area (a.u.)	143	190	167	176	155	166	16.3	9.8
Wt. norm. antioxidant peak area (a.u.)	771	996	771	940	877	871	89.9	10.3
I.S. corrected antioxidant peak area	1024	996	877	1014	1075	997	65.6	6.6

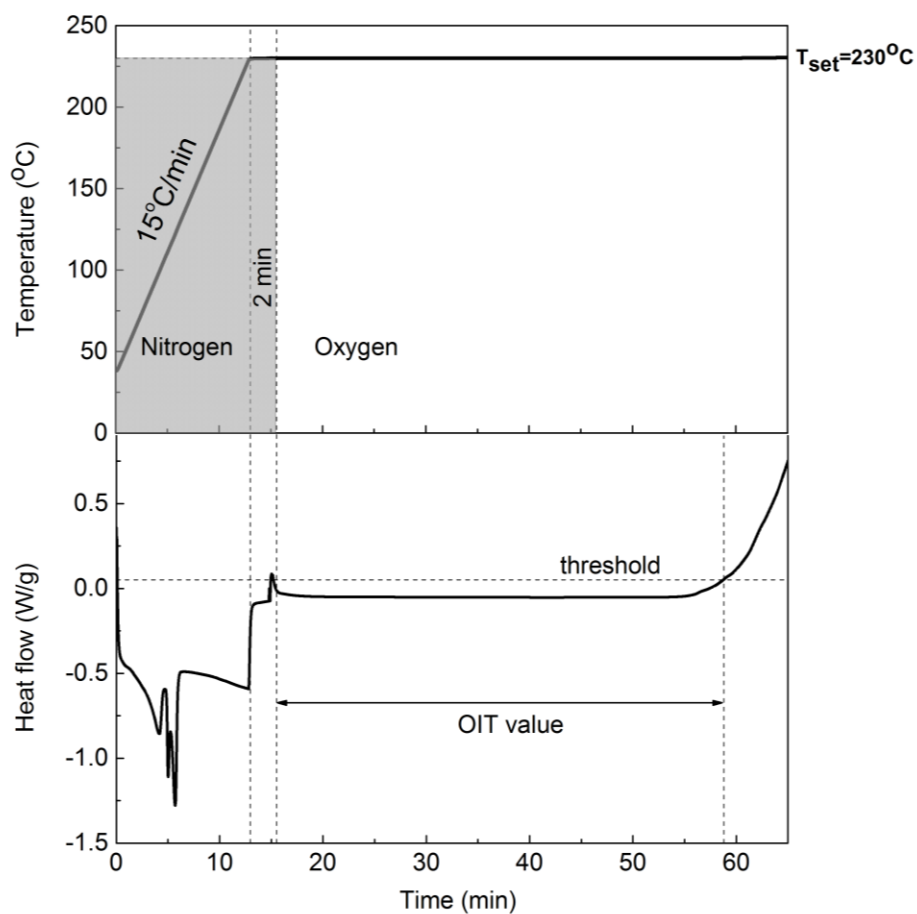


Figure 2. Temperature and gas profile and corresponding heat flow of OIT measurement. The example heat flow curve shown here is for pristine XLPE sample material.

Results

Figure 3 shows the evolved gas temperature profiles of a) the material as a whole, b) the XLPE polymer matrix, c) the brominated flame retardants, and d) the antioxidant. In Figure 3 a), it is shown that gases start to evolve from the test piece at around 150 °C, and this evolution continues until the ultimate temperature 800 °C of the pyrolyzer temperature program. Figures 3 b), c), and d) show that the XLPE polymer matrix starts to decompose between 300 and 400 °C and the majority of the polymer pyrolysis occurs between 400 and 500 °C. The brominated flame retardants show significant desorption and decomposition starting between 300 and 400 °C. The decomposition of antioxidant poly(1,2-dihydro-2,2,4-trimethylquinoline) (pTMQ) takes place mainly in the temperature range 200 to 350 °C.

Of the 13 sample scenarios studied in this work, the spectra of selected samples are displayed in Figures 4 and 5. Other spectra can be found in [15]. Figure 4 displays the mass spectra of samples that were aged at 60 °C for 15 days with gamma radiation exposure at doses of 0, 45, 110, and 190 kGy. Figure 5 displays the mass spectra of samples that were thermally-only aged for 15 days at three different temperatures of 60, 90, and 115 °C. Main peaks of the antioxidant are marked with arrows in the spectrum of the pristine sample in Figure 4. It is observed that the height of antioxidant peaks decrease with increasing gamma radiation exposure and increasing aging temperature. Peak areas were calculated and the antioxidant quantities in pristine and different samples, normalized to the antioxidant content of the pristine sample, are plotted in Figure 6. At the same aging temperature, the antioxidant content decreases with higher gamma radiation dose. At the same radiation dose, the antioxidant content decreases with higher aging temperature. The sample that was aged at 115 °C with exposure to gamma radiation at the dose of 190 kGy showed maximum depletion (93 %) in the quantity of antioxidant. The sample that was aged at 60 °C without gamma radiation showed minimum depletion (7 %) in the quantity of antioxidant. The depletion of antioxidant of other samples lies in between 7 % and 93 %.

Figure 7 shows the measured OIT values of the same sample set. At the same aging temperature, the OIT decreases with higher gamma radiation dose. At the same radiation dose, the OIT decreases with higher aging temperature. The sample that was aged at 115 °C with exposure to gamma radiation at the dose of 190 kGy showed the maximum reduction of 80 % in OIT. The sample that was aged at 60 °C without gamma radiation showed no reduction in OIT. Figure 8 compares normalized antioxidant quantity and normalized OIT of the sample set.

Discussion

Generally speaking, the depletion of antioxidant may be caused by evaporation, sublimation, and chemical reactions with free radicals. Prior to further discussion on how and why the changes in antioxidant pTMQ vary in differently aged samples, the possible chemical reaction mechanisms of pTMQ with free radicals generated in the aging process are discussed as follows.

Most oxidative mechanisms characteristic of the degradation of polymers during processing, storage and long-term end-use involve free radicals induced thermally, catalytically, mechano-chemically, or by radiation [21]. Reactions (1) through (4) show one chain of reactions that may take place in the polymer degradation process without the involvement of antioxidant [22]. Bond scission causes the generation of free radicals R^\bullet , as shown in (1), which, in the presence of oxygen, react to produce more unstable free radicals as the oxidation process repeats itself in the polymer, as shown in (2), (3), and (4). The process in (1) is termed the degradation *initiation*, and the ensuing processes fall under

degradation *propagation*. Degradation propagation processes take place at a much faster rate than degradation initiation [22].

The antioxidant used in the studied XLPE insulator is pTMQ, which is a type of hindered heterocyclic aminic stabilizer. The most widely used aminic stabilizers are grouped into two categories. One consists of aromatic and non-hindered heterocyclic amines, the classical antidegradants applied mainly in rubbers and to a lesser extent in polyolefins as antioxidants, heat stabilizers, antifatigue agents and antiozonant [23, 24]. The second group consists of hindered heterocyclic amines introduced on the market in the 1970s. These rank among the currently most extensively studied additives [21]. Hindered amine stabilizers have been utilized almost exclusively in polyolefins and coatings as light stabilizers, photo-antioxidants, and heat stabilizers. The antioxidant pTMQ belongs to the second group, hindered heterocyclic aminic stabilizers, and efficiently stabilizes XLPE used as insulation material for electric cables exposed to irradiation doses [25]. Generally speaking, interference with free-radical and peroxidic species resulting in polymer autoxidation, fatigue and photo-oxidation belong to the principal processes of polymer stabilization [21]. The detailed discussion of reaction mechanisms of various aminic stabilizers can be found in [24, 26, 27]. Specifically, pTMQ has a broad application spectrum. It has been used for a long time as a heat stabilizer and antioxidant with some anti-flex-crack effect in the rubber industry and in polyolefins [28]. pTMQ is generally more efficient than other common antioxidants in protecting against irradiation aging and thermal oxidation below 150 °C [29]. Abstraction of the hydrogen atom from the –NH group of TMQ according to Reaction (5) is the primary step of the antioxidant action [21].

When the samples are aged thermally, without gamma radiation, C-C bond breakage is most likely caused by thermal fluctuations, and the C-C bond breakage leads to the generation of free radicals. We speculate that the rate of generation of free radicals increases with increasing aging temperature. In the presence of gamma radiation, however, the rate of generation of free radicals is likely to be much higher than when gamma radiation is absent, due to the high photon energy of gamma radiation. The C-C bond energy is about 348 kJ/mol [30], and the photon energy from Co-60 radiation source is 1.17 MeV (1.129×10^8 kJ/mol) and 1.33 MeV (1.283×10^8 kJ/mol) [31], easily capable of breaking C-C bonds and generating free radicals. Moreover, the high energy photons generate secondary electrons which break additional polymer C-C bonds, producing more free radicals.

The melting temperature of pTMQ lies in the range from 72 to 94 °C [32], and its boiling temperature is higher than 315 °C [32]. When thermal aging takes place at 60 °C, sublimation and rate-limited chemical reactions might take place, causing depletion by only 15% as observed in Figure 6. When thermal aging takes place at 90 and 115 °C, evaporation, sublimation and rate-enabled chemical reaction take place simultaneously, causing a much faster depletion rate and explaining the observed depletion of approximately 60 and 70 % in these cases, respectively. Adding gamma radiation to the exposure environment causes the depletion of pTMQ to occur at a much faster rate, likely due to the faster generation of polymer free radicals as described in the previous paragraph.

The normalized values of OIT and antioxidant content plotted together in Figure 8 show dramatic changes in antioxidant content for all but the two most lightly aged samples (pristine and 60C-0). Figure 8 also reveals a dramatic reduction in antioxidant content, more than 50 %, before any significant change in OIT is observed. The samples that have lost more than 50 % of antioxidant (i.e. all but the two most lightly aged samples) display a linear relation between OIT and normalized antioxidant content, suggesting that OIT could be used as an indicator of age-induced antioxidant depletion in XLPE-based cable insulation materials after 50 % of antioxidant has been lost. Depletion of antioxidant may not be the only factor that contributes to the change in OIT in the samples studied. Besides depletion of antioxidant, OIT may also be affected by the polymer type, polymer structure,

and other additives and fillers. Scission of XLPE polymer chains and alteration of the crystalline and semi-crystalline content of the XLPE may lead to reduction in OIT in addition to changes caused by consumption of antioxidant.

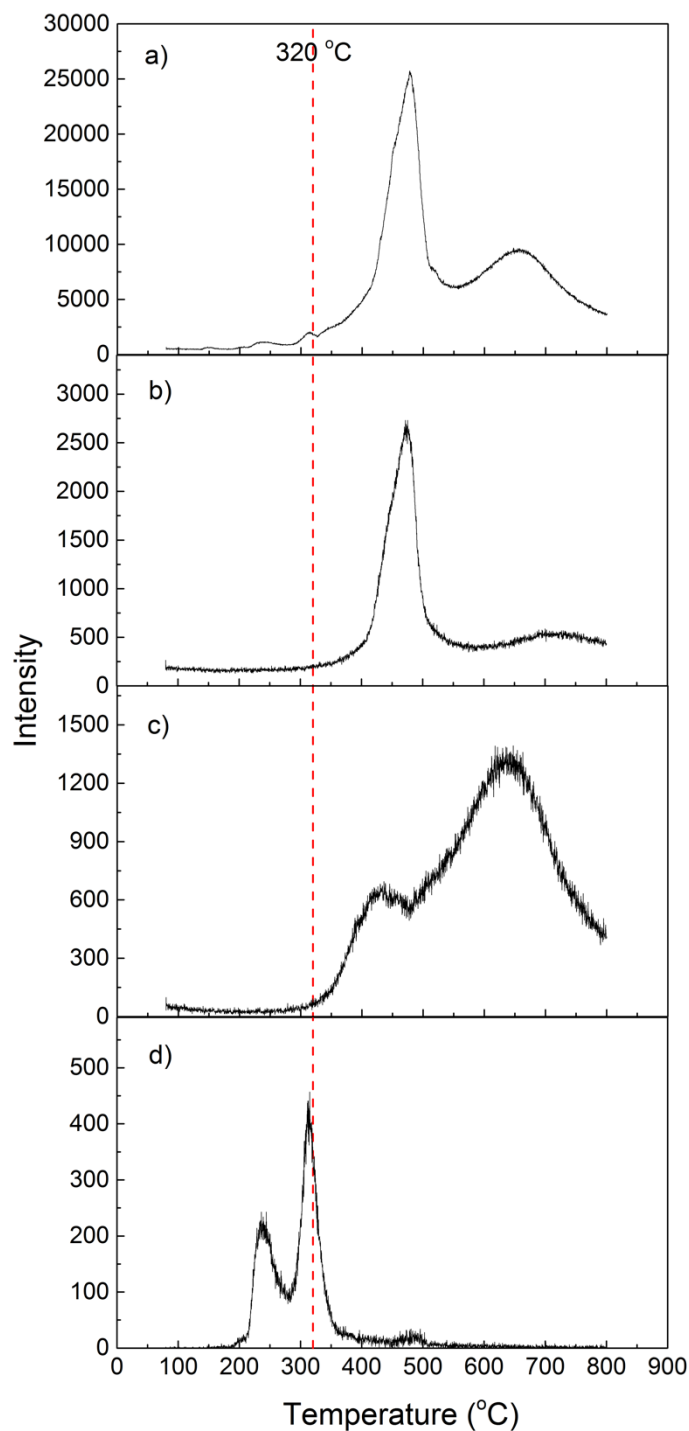


Figure 3. Evolved temperature profiles of major components in XLPE-based cable insulation material; a) evolved pyrolyzates from the whole sample piece; b) selected evolved pyrolyzates of XLPE polymer; c) evolved pyrolyzates from brominated flame retardants; and d) evolved pyrolyzates from antioxidant.

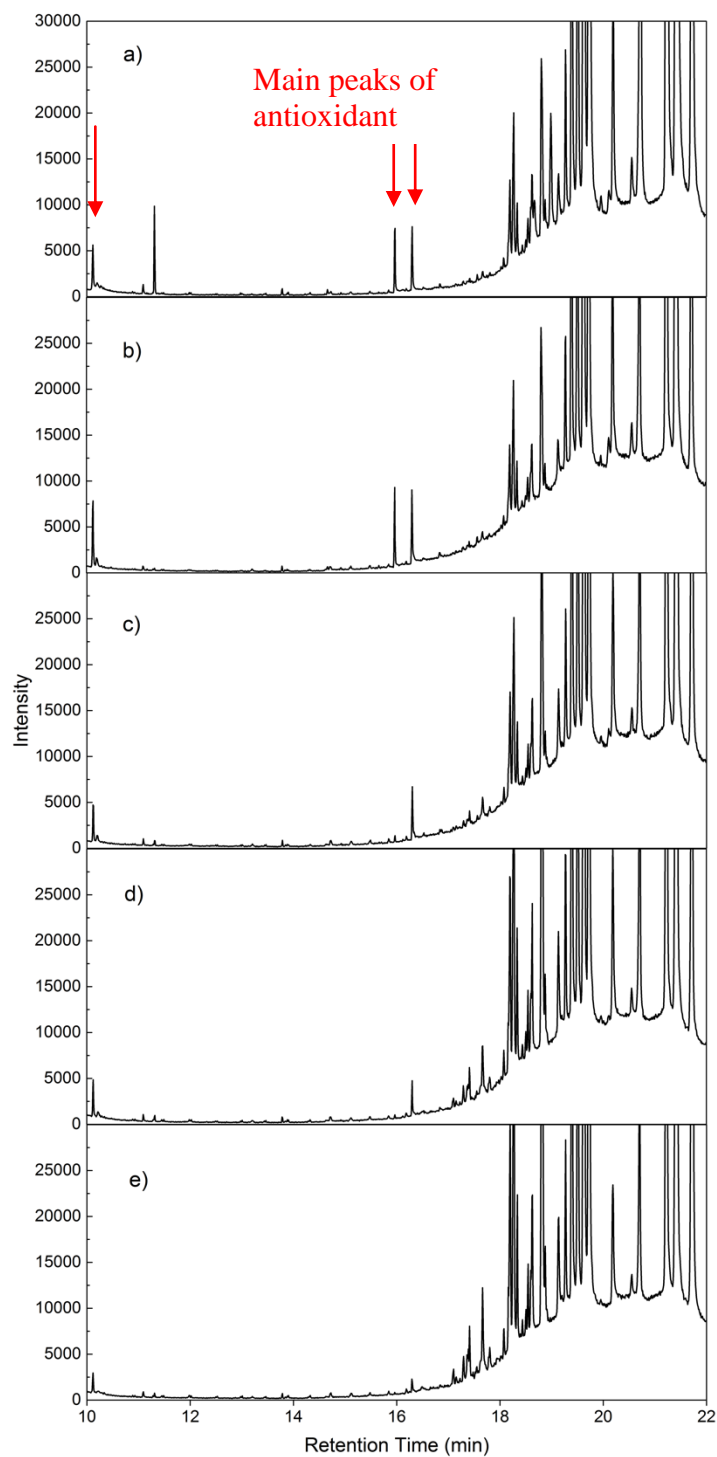


Figure 4. Mass spectra of XLPE-based samples a) pristine, and those that have been simultaneously thermal and gamma radiation aged at 60 °C for 15 days, with gamma radiation exposure of b) 0, c) 45, d) 110, and e) 190 kGy. Main peaks of the antioxidant are marked with arrows in the spectrum of the pristine sample. The peak at retention time of ~ 11.5 min that appears only in spectrum a) indicates the presence of residual cross-linking agent.

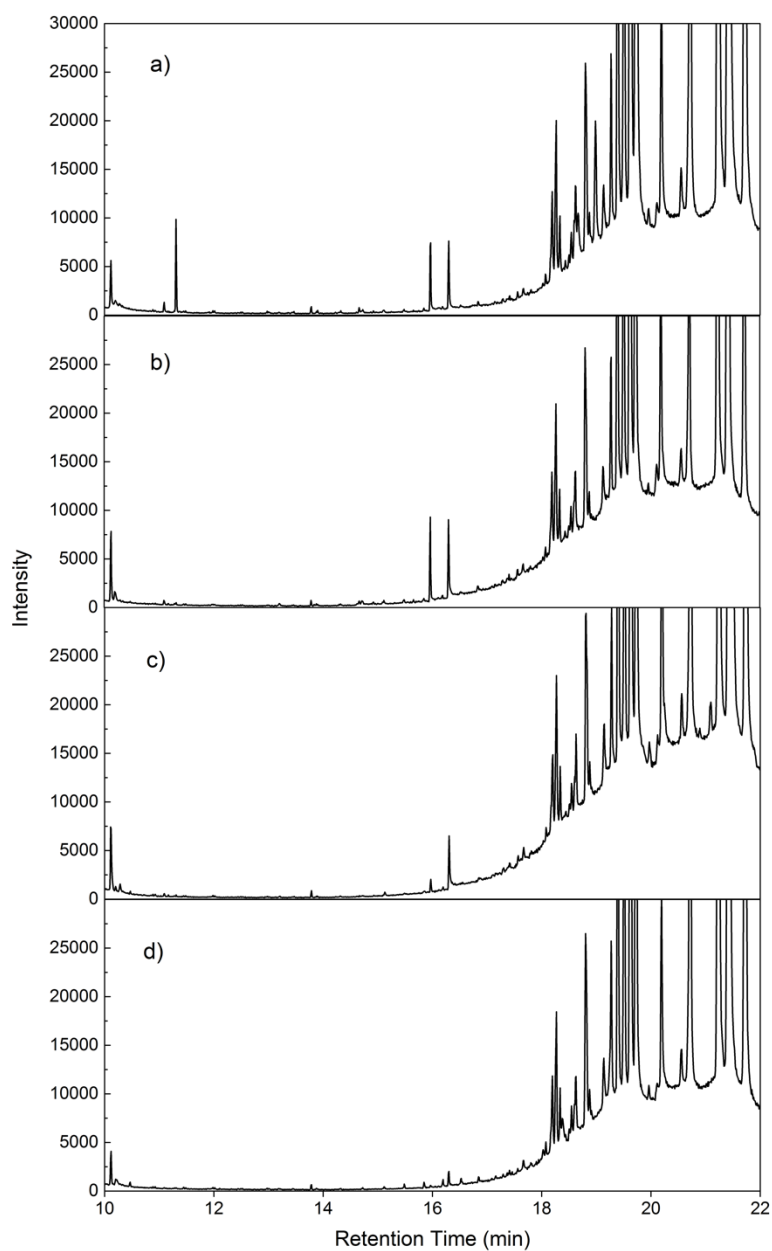


Figure 5. Mass spectra of XLPE-based samples a) pristine, and those that have been thermally aged at b) 60, c) 90, and d) 115 °C for 15 days, without gamma irradiation. The peak at retention time of ~ 11.5 min that appears only in spectrum a) indicates the presence of residual cross-linking agent.

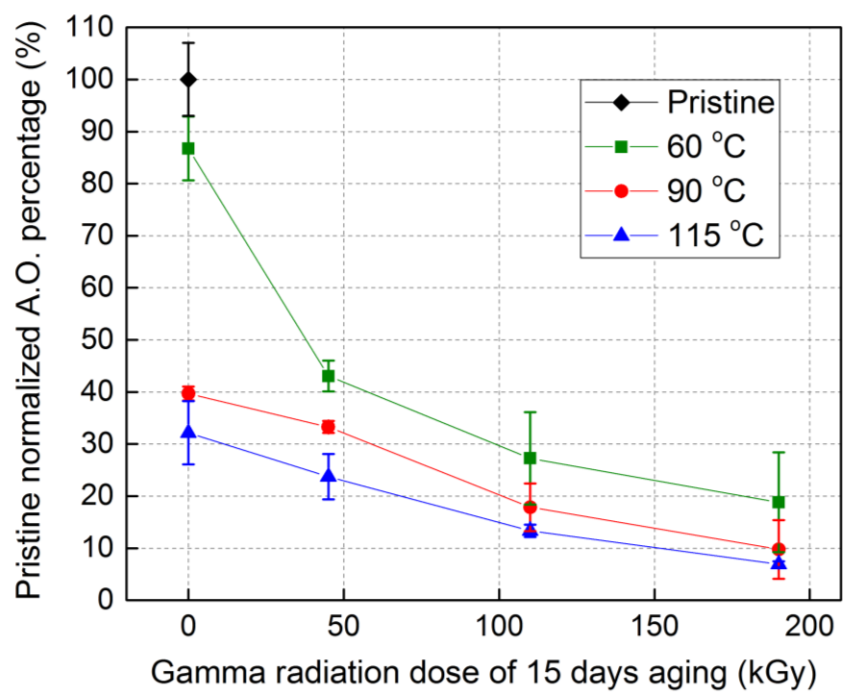


Figure 6. Antioxidant quantity in pristine and aged XLPE-based samples, normalized to the value for the pristine sample (997 area units).

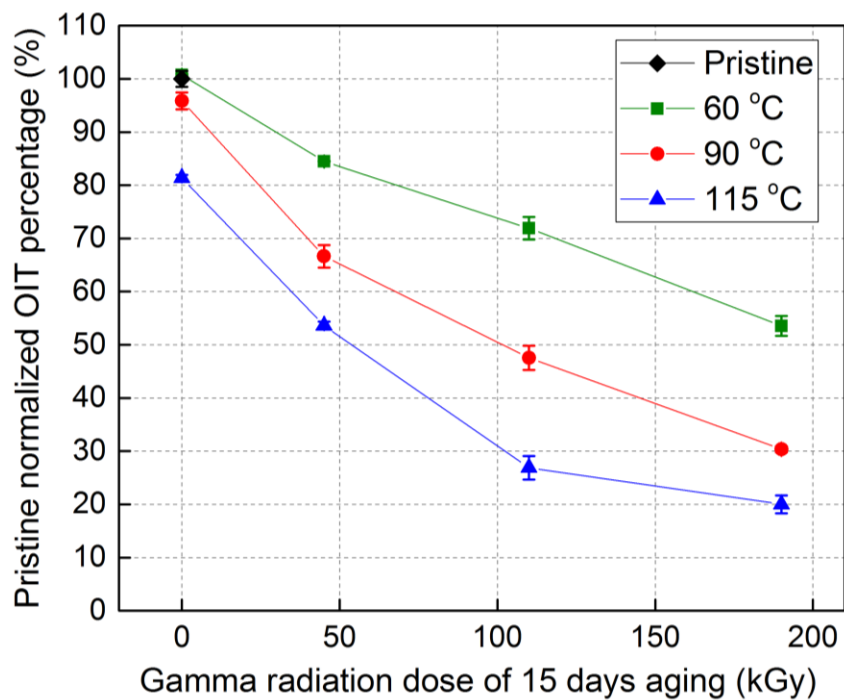


Figure 7. OIT for pristine and aged XLPE-based samples, normalized to the value for the pristine samples (48.6 min).

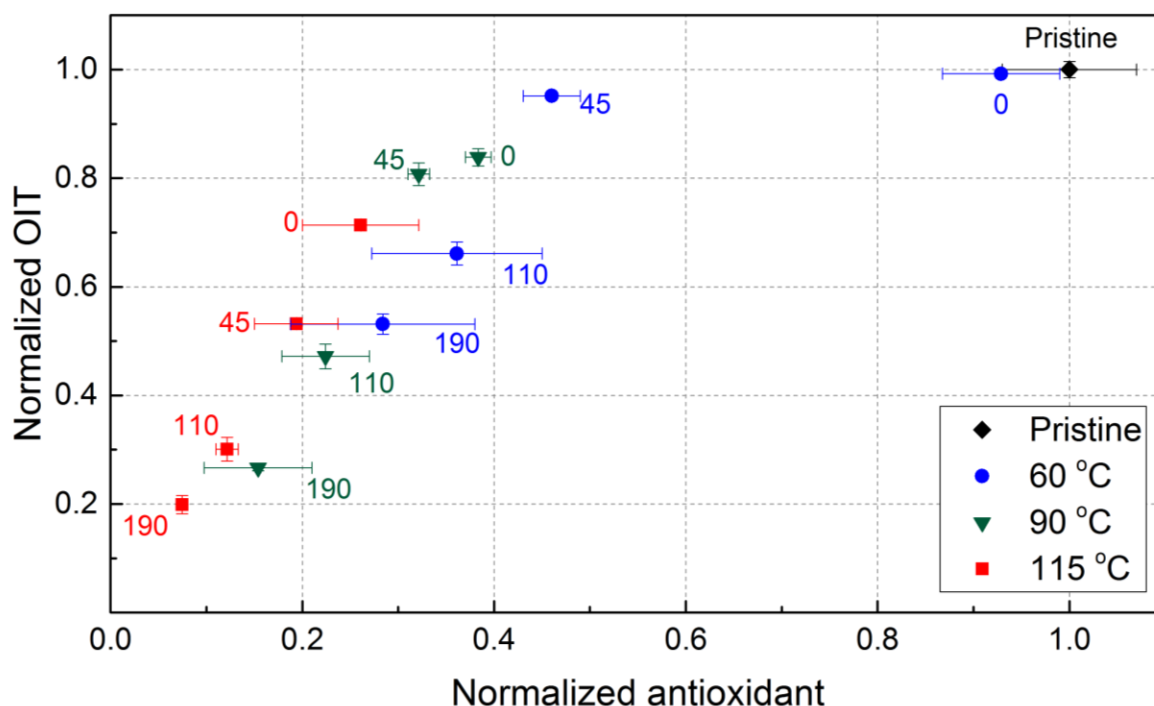
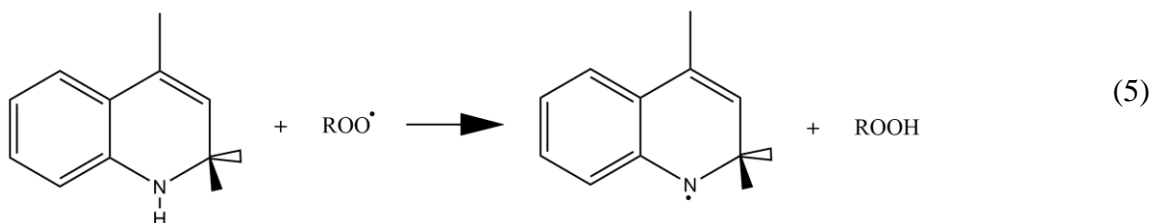
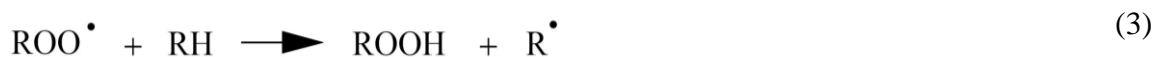


Figure 8. OIT and antioxidant content normalized to values for the pristine sample. The numbers 0, 45, 110, and 190 indicate gamma radiation dose in kGy.



Conclusion

Quantitative analysis of antioxidant in a commercially-available, cross-linked polyethylene-based cable insulation material for nuclear power plant applications has been performed. Comparison between the antioxidant quantity in the material and the oxidation induction time (OIT) of the material

was made. For isothermal aging, the antioxidant quantity and OIT were observed to decrease with increasing gamma radiation dose. At fixed gamma radiation dose, they were observed to decrease with increasing aging temperature. The depletion of antioxidant quantity ranges from 7 % to 93 %, and the decrease of OIT ranges from 0 to 80 % for samples studied in this paper. After initial loss of approximately 50 % of the antioxidant, the changes in the quantity of antioxidant in the material follow the same trend as the changes in OIT of the material when aged under various conditions. Therefore OIT can be an effective indicator of the antioxidant depletion severity in XLPE based cable insulation materials, for depletion levels greater than 50 %, while depletion of antioxidant is not the only factor that contributes to the reduction in OIT of the studied material. The way in which the antioxidant may slow the degradation process of the polymer, by consuming the majority of the free radicals generated in the initial stages of degradation, has been described.

Acknowledgement

This work was funded by the DOE Office of Nuclear Energy's Nuclear Energy University Programs under contract number DENE0008269 and the DOE Office of Nuclear Energy's Light Water Reactor Sustainability Program. Exposure experiments were conducted at Pacific Northwest National Laboratory, which is operated by Battelle for the US DOE under contract DE-AC05-76RL01830. The authors acknowledge useful discussion with Andrew C. Kolbert, CTO of Avomeen Analytical Services, LLC.

References

- [1] U.S. Energy Information Administration, How many nuclear power plants are in the united states, and where are they located?, 2017, August 15. <https://www.eia.gov/tools/faqs/faq.php?id=207&t=3>.
- [2] World Nuclear Association, Nuclear power reactors, 2018, January. <http://www.world-nuclear.org/information-library/nuclear-fuel-cycle/nuclear-power-reactors/nuclear-power-reactors.aspx>.
- [3] M. Subudhi, R. Lofaro, U.S.N.R.C.O.o.N.R.R.D.o.E. Technology, L. Brookhaven National, Literature review of environmental qualification of safety-related electric cables, Washington, DC : Division of Engineering Technology, Office of Nuclear Regulatory Research, U.S. Nuclear Regulatory Commission ; Supt. of Docs., U.S. G.P.O., distributor, Washington, DC, 1996.
- [4] N. Bowler, S. Liu, Aging Mechanisms and Monitoring of Cable Polymers, International Journal of Prognostics and Health Management, 2015.
- [5] H.A. Khonakdar, S.H. Jafari, S. Rasouli, J. Morshedien, H. Abedini, Investigation and Modeling of Temperature Dependence Recovery Behavior of Shape-Memory Crosslinked Polyethylene, Macromol. Theory Simul. 16(1) (2007) 43-52.
- [6] I. Chodák, Properties of crosslinked polyolefin-based materials, Progress in Polymer Science 20(6) (1995) 1165-1199.
- [7] M. Palmlöf, T. Hjertberg, Crosslinking of poly(ethylene- co-1,9-decadiene) by electron beam irradiation, Polymer 41(17) (2000) 6481-6495.
- [8] L. Bustard, P. Holzman, Low-voltage environmentally-qualified cable license renewal industry report: Revision 1. Final report, ; Electric Power Research Inst., Palo Alto, CA (United States); Sandia National Labs., Albuquerque, NM (United States); Strategic Technology and Resources, Winchester, MA (United States), 1994.
- [9] F. Gaston, N. Dupuy, S.R.A. Marque, D. Giges, S. Dorey, Monitoring of the discoloration on [gamma]-irradiated PE and EVA films to evaluate antioxidant stability.(Report), Journal of Applied Polymer Science 135(18) (2018).

- [10] H. Wang, L. Li, J. Guan, H. Jiang, R. Shen, X. Ding, J. Li, Y. Li, Investigation on Molecular Structures of Electron-Beam-Irradiated Low-Density Polyethylene by Rheology Measurements, *Ind. Eng. Chem. Res.* 57(12) (2018) 4298-4310.
- [11] B. Bartoníček, V. Plaček, V. Hnát, Comparison of degradation effects induced by gamma radiation and electron beam radiation in two cable jacketing materials, *Radiation Physics and Chemistry* 76(5) (2007) 857-863.
- [12] D. Jeon, G. Park, I. Kwak, K. Lee, H. Park, Antioxidants and their migration into food simulants on irradiated LLDPE film, *LWT-Food Sci. Technol.* 40(1) (2007) 151-156.
- [13] R. Mortimer, T. Varley, Quantification of colour stimuli through the calculation of CIE chromaticity coordinates and luminance data for application to in situ colorimetry studies of electrochromic materials, *Displays* 32(1) (2011) 35-44.
- [14] F. Bourges, G. Bureau, J. Dumonceau, B. Pascat, Effects of electron beam irradiation on antioxidants in commercial polyolefins: Determination and quantification of products formed, *Packaging Technology and Science* 5(4) (1992) 205-209.
- [15] S. Liu, Composition identification, aging mechanisms and nondestructive aging indicator of commercial filled cross-linked polyethylene (XLPE) cable insulation materials, Iowa State University Digital Repository, 2017.
- [16] L. Shuaishuai, L.S. Fifield, N. Bowler, Towards aging mechanisms of cross-linked polyethylene (XLPE) cable insulation materials in nuclear power plants, 2016 IEEE Conference on Electrical Insulation and Dielectric Phenomena (CEIDP), 2016, pp. 935-938.
- [17] Y.G. Hsuan, M. Li, Temperature and pressure effects on the oxidation of high-density polyethylene geogrids, *Geotextiles and Geomembranes* 23(1) (2005) 55-75.
- [18] W. Mueller, I. Jakob, Oxidative resistance of high-density polyethylene geomembranes, *Polymer Degradation and Stability* 79(1) (2003) 161-172.
- [19] M. Uhniat, S. Kudła, Stabilisation of LDPE cross-linked in the presence of peroxides I. Kinetic study of the oxidation, *Polymer Degradation and Stability* 71(1) (2000) 69-74.
- [20] L.S. Fifield, L. Shuaishuai, N. Bowler, Simultaneous thermal and gamma radiation aging of cable polymers, 2016 IEEE Conference on Electrical Insulation and Dielectric Phenomena (CEIDP), 2016, pp. 11-14.
- [21] J. Pospíšil, Aromatic and heterocyclic amines in polymer stabilization, *Polysoaps/Stabilizers/Nitrogen-15 NMR*, Springer Berlin Heidelberg, Berlin, Heidelberg, 1995, pp. 87-189.
- [22] Nocil Limited, Antioxidants and antidegradants.
<http://www.nocil.com/Downloadfile/ETechnicalNote-Antioxidants-Dec2010.pdf>.
- [23] J. Pospíšil, P.P. Klemchuk, Oxidation inhibition in organic materials / editors, Jan Pospíšil, Peter P. Klemchuk, Boca Raton, Fla. : CRC Press, Boca Raton, Fla., 1990.
- [24] J. Pospíšil, Functionalized oligomers and polymers as stabilizers for conventional polymers, *Polymer Compositions Stabilizers/Curing*, Springer Berlin Heidelberg, Berlin, Heidelberg, 1991, pp. 65-167.
- [25] J.W. Ray, A.B. Reynolds, Effect of Antioxidants on Radiation Stability of Polymers Used for Electric Cable Insulation, *Nuclear Technology* 91(3) (1990) 394-403.
- [26] E.T. Denisov, I.V. Khudyakov, Mechanisms of action and reactivities of the free radicals of inhibitors, *Chemical Reviews* 87(6) (1987) 1313-1357.
- [27] Z. H., Polymer Stabilization and Degradation. Hg. von P. P. Klemchuk. ACS Symposium Series 280., *Acta Polymerica* 38(1) (1987) 101-102.
- [28] P. M., Rubber technology handbook, *British Polymer Journal* 23(4) (1990) 359-359.
- [29] E. Richaud, X. Colin, C. Monchy-Leroy, L. Audouin, J. Verdu, Polyethylene stabilization against thermal oxidation by a trimethylquinoleine oligomer, *Polymer Degradation and Stability* 94(3) (2009) 410-420.

- [30] J.C. Earl, Carbon-carbon bond energies, Tetrahedron 9(1) (1960) 65-66.
- [31] J.K. Shultis, R.E. Faw, Fundamentals of nuclear science and engineering, New York : Marcel Dekker, New York, 2002.
- [32] Chemical Book, Poly(1,2-dihydro-2,2,4-trimethylquinoline) 2017.
https://www.chemicalbook.com/ChemicalProductProperty_EN_CB3222852.htm.