Predictive Model on the Reliability of the Insulation Made from Special Heat-Resistant Polyvinyl Chloride

Yuan-Shang Chang, Yizhen Zhang, and Ali Mosleh
B. John Garrick Institute for the Risk Sciences, and Department of Materials Science & Engineering
University of California, Los Angeles (UCLA), USA

Abstract: Cables are exposed to radiation and heat in nuclear power plants (NPPs). These two degradation factors cause the brittleness of the cable insulation, which may render the exposure of the metallic core in a cable. The degree of the brittleness of the insulation is often characterized by the elongation at break (EAB) measured by a tensile test. Due to the safety concern of NPPs, developing a model predicting the EAB as a function of time becomes an important topic. Starting with reaction kinetics, we have developed the Dichotomy Model to model the decrease of the EAB of the cable insulation made from cross-linked polyethylene (XLPE or PEX), ethylene propylene rubber (EPR), and silicone rubber (SIR). To demonstrate the generic nature of this newly-proposed model, the experimental data of another commonly-used insulation material, special heat-resistant polyvinyl chloride (SHPVC), is used to validate the model. SHPVC specimens were simultaneously exposed to radiation and heat, and the EAB value versus time was measured. Both deterministic and probabilistic approaches are included in this research to quantify the reliability of the insulation. The modeled results fit the experimental data, showing that the Dichotomy Model is unlimited to specific insulations.

Keywords: Cable insulation, thermal degradation, Bayesian parameter estimation, Dichotomy Model, special heat-resistant polyvinyl chloride (SHPVC), radiation

1. INTRODUCTION

Cables transmit power and signals in nuclear power plants. The insulations of the cables degrade due to heat and radiation. Degraded insulations become brittle. When external forces are exerted, cracks tend to form on the surface of an insulation [1], which may lead to the exposure of the metallic core in a cable. The brittleness of cable insulation is widely characterized by the elongation at break (EAB) via a tensile test [2-5]. Therefore, the degree of the degradation of the insulation is often quantified by the EAB in the field application [6-9].

There are four major polymer materials for cable insulations: cross-linked polyethylene (XLPE or PEX), ethylene propylene (EPR), silicone rubber (SIR), and polyvinyl chloride (PVC) [9]. To predict the decrease of their EAB as time elapses, two models had been developed in the literature. The first model is Time-temperature superposition (TTS) [4, 10]. TTS is also known as Time to Equivalent Damage or Superposition of Time-Dependent Data [4, 9-11]. This model has been used for more than 50 years [4]. However, it cannot accommodate the change of the shape of the EAB curve against time, though the change of the shape often occurs when aging conditions are varied [12]. The second model is Dose to Equivalent Damage (DED) [4]. This approach focuses on the prediction of a specific EAB value. In other words, an EAB vs. time curve is unavailable when DED is applied.

In our previous study [13-15], we have developed Dichotomy Model, which can accommodate the change of the shape of an EAB curve plotted versus time with respect to varied aging conditions. Different from DED model focusing on one EAB value, the continuous values of the EAB as a function of time is formulized in Dichotomy Model. This model has been applied to the degradation of the insulations made from XLPE, EPR, and SIR [13-15]. To demonstrate the generic nature of the proposed model, in this paper, we have applied this model to the radiation and thermal degradation of another insulation material: special heat-resistant polyvinyl chloride (SHPVC), which has not been done in the literature. In addition to developing a deterministic approach calculating the time-to-failure of the cable insulation by Dichotomy Model, a probabilistic approach [16-20] based on Bayesian parameter estimation has also been derived in this paper to represent the reliability of the insulation with uncertainty.

2. MODELING

When SHPVC insulation is exposed to gamma radiation emitted from Cobalt 60, the trend of the EAB against time (t) [9] can be schematically represented by the solid line in Figure 1. At the early stage of the degradation, the drop-off rate of the EAB is insignificant. Therefore, in our model, the solid line before τ_0 can be represented by a horizontal line denoted by the dotted line. τ_0 is the length of this horizontal section and named incubation time [13-15]. After τ_0, the drop-off rate of the EAB increases. Based on the Dichotomy Model, the solid line between τ_0 and τ_1 can be modeled by equation (1) [13-15], which is plotted by the dashed line. In equation (2), V_d is degradation ratio with no unit; ν is the drop-off rate of the EAB against time in the unit of [1/time], which is assumed constant in this study. When the aging time is longer than τ_1, the degradation accelerates. While the minimum of the EAB for the field application is above the value denoted by the chained line in Figure 1, no model is needed for the solid line after τ_1.

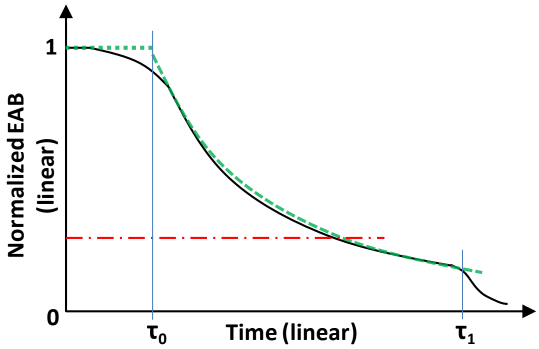

Figure 1. The trend of the EAB of SHPVC

$$\text{Normalized EAB} \equiv \kappa \cong 1 - (V_d)^{(1/3)} \tag{1}$$

where

$$V_d = 1 - e^{-\nu(t-\tau_0)} \tag{2}$$

or

$$\kappa \cong 1 - \left\{1 - e^{-\nu(t-\tau_0)}\right\}^{\left(\frac{1}{3}\right)} \tag{3}$$

Least square is a method that can determine the values of ν and τ_0 by making the dashed line plotted by equation (3) fit the solid between τ_0 and τ_1. However, there are always differences between the predicted results represented by the dashed line and the experimental data denoted by the solid line. Equation (4) is the concept of Bayesian parameter estimation, which can determine the distribution of ν. By plugging this distribution into equation (3), the dashed line becomes a band defining the level of the confidence with respect to predicted value.

$$\Pr(\theta \mid \text{data}) = \frac{\Pr(\text{data} \mid \theta)\Pr(\theta)}{\int \Pr(\text{data} \mid \theta)\Pr(\theta)d\theta} \tag{4}$$

To develop the likelihood function of equation (4), it is reasonable to assume that the measured data (κ) are normally distributed around a predicted value (μ_κ) with a standard deviation σ. That is:

$$\kappa \sim \mathcal{N}(\mu_\kappa, \sigma) \tag{5}$$

where μ_κ can be calculated by equation (3) and represented by equation (6).

$$\mu_\kappa = 1 - [1 - \exp(-\nu \times (t - \tau_0))]^{1/3} \tag{6}$$

With respect to normal distribution and based on equation (6), the likelihood function of equation (4) can be represented by equation (7) where the priors are ν and σ for Bayesian parameter estimation.

$$\Pr(\kappa, t|\nu, \sigma) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left(-\frac{\left(\kappa - (1 - [1 - \exp(-\nu \times (t - \tau_0))]^{\frac{1}{3}})\right)^2}{2\sigma^2} \right) \tag{7}$$

After applying equation (7) to equation (4), the distribution of ν can be determined. These approaches render the uncertainty of the EAB versus time based on equation (3).

3. VALIDATION

The experimental data of the SHPVC made by company A reported in the reference [9] are used to validate the proposed model. The dose rate is about 100 Gy/h and the aging temperatures are 80°C, 90°C, and 100°C, respectively. Measured EAB against time is plotted by the discrete patterns in Figure 2, Figure 4, and Figure 6. Least square is applied to equation (3) to determine the values of τ_0 by testing different values of τ_0 and ν. The purpose is to make the equation (3) fit the experimental data. The values of the τ_0 determined by this approach are listed in the captions of Table 1, Table 2, and Table 3 with respect to different aging conditions. Determined by least square, the values of the ν corresponding to 80°C, 90°C, and 100°C are 2.424×10^{-4} [1/h], 2.423×10^{-4} [1/h], and 3.343×10^{-4} [1/h], respectively.

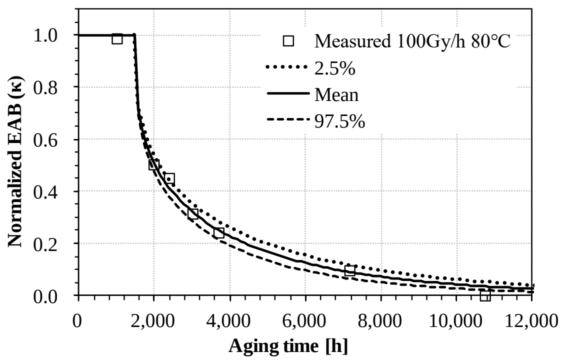

Figure 2. Measured (the discrete pattern) and modeled (continuous lines) EAB at 100Gy/h 80°C

To increase the precision of the prediction, measured EAB values before τ_0 or after τ_1 shall be excluded. For the experimental data at 80°C, experimental data before 1,400 hours or after 8,000 hours are excluded when equation (7) is applied. The priors, ν and σ, are assumed uniform distributions ranging from 0 to 0.0004 and from 0 to 0.5, respectively. The results of Bayesian parameter estimation are plotted in Figure 3, showing the distribution of ν and σ. The percentile of the distributions is listed in Table 1. The dotted line in Figure 2 is plotted by equation (3) with the ν at 2.5 percentile; the dashed line is plotted by the same equation with the ν at 97.5 percentile.

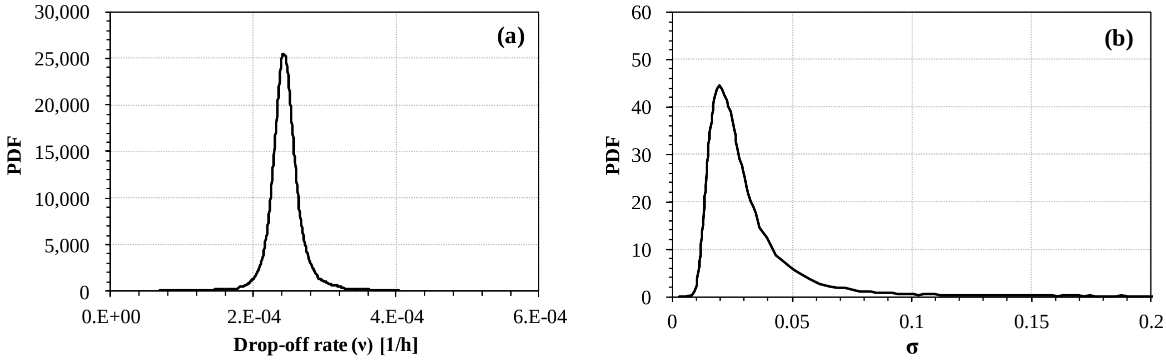

Figure 3. PDF of ν and σ at 100Gy/h and 80°C

Table 1. Drop-off rate (ν) at 80°C where τ_0 = 1,475 hours

Mean	2.5%	25%	50%	75%	97.5%
2.453	2.051	2.332	2.437	2.551	2.973

*Unit = [10^{-4}/h]

As for the experimental data at 90°C in Figure 4, the same approach is performed. Measured EAB before 1,400 hours or after 8,000 hours is excluded. The priors, ν and σ, are both assumed uniform distributions ranging from 0 to 0.0007 and from 0 to 0.5, respectively. After Bayesian parameter estimation, the distributions of ν and σ are plotted in Figure 5. The percentile corresponding to Figure 5 is listed in Table 2. The 2.5 and 97.5 percentile of the ν values are plugged into equation (3) respectively denoted by the dotted and dashed lines in Figure 4, which shows the uncertainty of the EAB against time in the radiation and thermal aging.

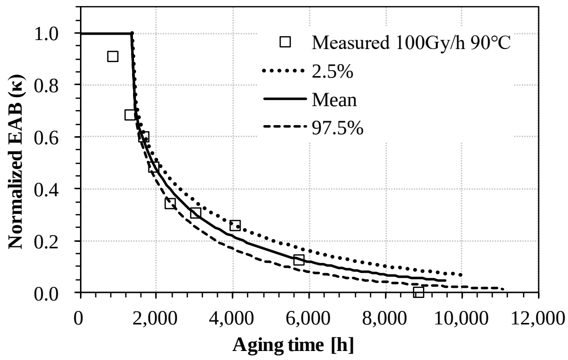

Figure 4. Measured (the discrete pattern) and modeled (continuous lines) EAB at 100Gy/h 90°C

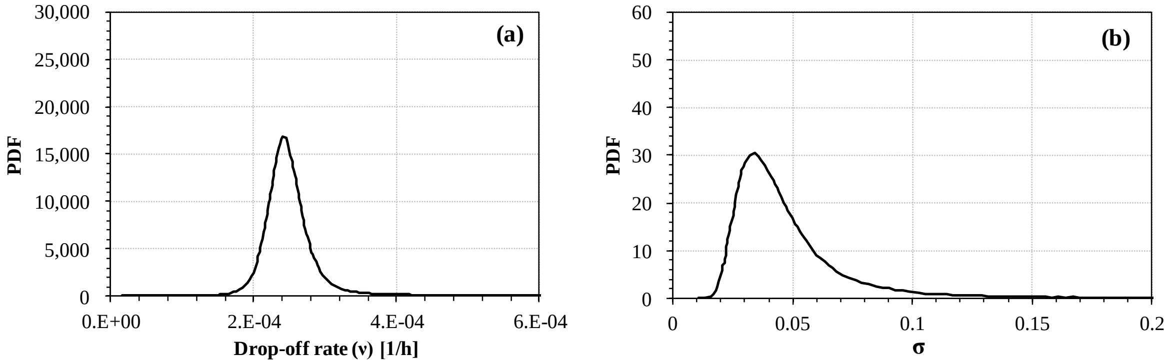

Figure 5. PDF of ν and σ at 100Gy/h and 90°C

Table 2. Drop-off rate (ν) at 90°C where τ_0 = 1,359 hours

Mean	2.5%	25%	50%	75%	97.5%
2.485	1.938	2.293	2.449	2.625	3.259

*Unit = [10^{-4}/h]

The same to the process applied to the experimental results at 80^{o}C and 90^{o}C, for the data at 100^{o}C plotted by the discrete pattern in Figure 6, the measured EAB before 1,400 hours or after 4,000 hours is excluded. The priors, ν and σ, are assumed uniform distributions ranging from 0 to 0.0007 and from 0 to 0.5, respectively. The distributions of ν and σ determined by Bayesian parameter estimation are shown in Figure 7. The percentile of Figure 7(a) is listed in Table 3. In the table, the values of ν at 2.5 and 97.5 percentile are plugged into equation (3), which is respectively represented by the dotted and dashed lines in Figure 6.

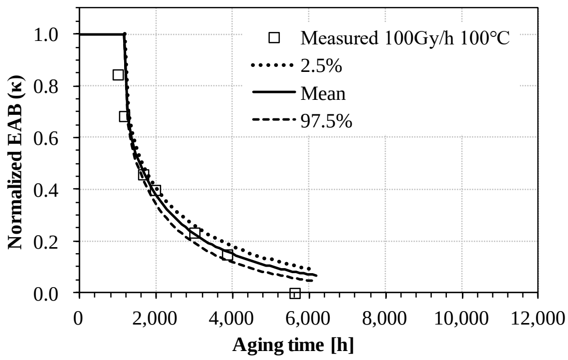

Figure 6. Measured (the discrete pattern) and modeled (continuous lines) EAB at 100Gy/h 100^{o}C

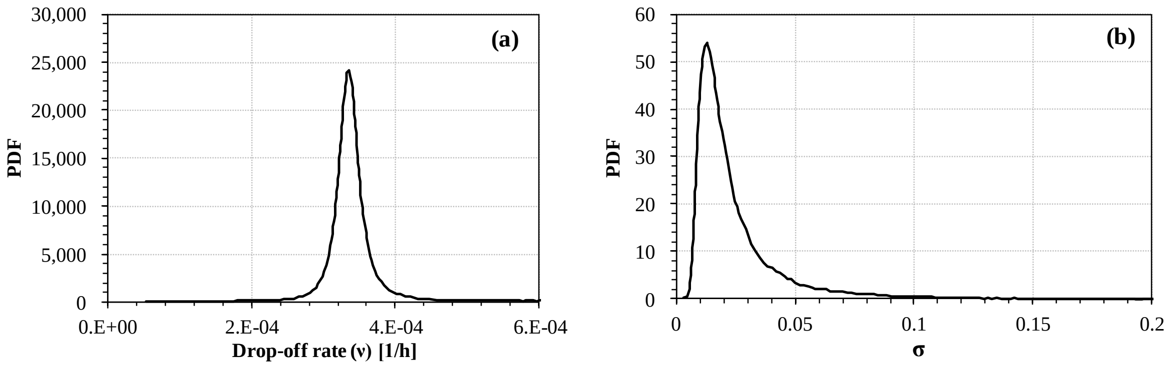

Figure 7. PDF of ν and σ at 100Gy/h and 100^{o}C

Table 3. Drop-off rate (ν) at 100^{o}C where τ_0 = 1,172 hours

Mean	2.5%	25%	50%	75%	97.5%
3.384	2.852	3.241	3.355	3.478	4.114

*Unit = [10^{-4}/h]

4. DISCUSSION

Degradation of PVC is caused mainly by dehydrochlorination process. Allylic chlorine and tertiary chlorine on a PVC chain are active sites for the process [21]. Besides, dehydrochlorination renders polyene structures on a PVC chain, which are initiators of dehydrochlorination. HCl generated by dehydrochlorination is a catalyst of the reaction itself. Therefore, HCl accelerates dehydrochlorination leading to chain scission, the decrease of the EAB, and the change of the color of PVC [22, 23]. When extensive dehydrochlorination is triggered by the catalyst among PVC chains, the trend of the EAB exceeds the domain of τ_0.

There are mainly three kinds of additives used in the field to prevent PVC from degrading: thermal stabilizers, antioxidants, and plasticizers. Thermal stabilizers can react with HCl and replace the active groups on PVC chains. It improves the weathering ability of PVC. In addition to thermal stabilizers, an antioxidant can also prevent PVC from aging. In PVC, heat and radiation generate free radicals [24-26] resulting in the degradation caused by oxidation. The antioxidant can slow the reaction rate of oxidation. A plasticizer is another kind of additive. It can prevent PVC from oxidizing as antioxidant does [27]. Moreover, plasticizers solvate PVC, which can improve the ductility of PVC by increasing the free volume between PVC chains [28]. In other words, plasticizers introduce intermolecular interactions such as van der Waals and dipole-dipole forces between PVC chains and plasticizers. Therefore, the loss of these three kinds of additives can lead to the decrease of the EAB.

Sufficient additives slow degradation rate and make the drop off of the EAB insignificant. This is one of the reasons for the existence of incubation time (τ_0) illustrated in Figure 1. When the aging time is longer than τ_0, the concentration of the additives decreases to a certain level due to the decomposition of the additives themselves, or the diffusion of the additives from the inside to the outside of the insulation. Insufficient additives can increase the degradation rate. Therefore, we model the trend of an EAB curve by two sections: before τ_0, ν is assumed negligible, while after τ_0, ν is determined by the Dichotomy Model. As for the aging time longer than τ_1, the additives may be depleted, so the drop-off rate becomes larger.

As shown in Figure 2, Figure 4, and Figure 6, the continuous lines plotted by equation (3) fit the experimental data, meaning that the proposed model is validated. There are two parameters in the model: ν and τ_0. Figure 8 (a) indicates that the values of ν insignificantly change when aging temperature is elevated from 80°C to 90°C, compared to the change from 90°C to 100°C. This is because the dose rate is at 100 Gy/h. In this high radiation environment, the degradation process is dominated by radiation effect. Therefore, the Arrhenius plot of Figure 8 (a), represented by Figure 8 (b), does not show a good linear fit. The second parameter, τ_0, possesses a similar trend to that of ν. Figure 9 shows that the difference of τ_0 between 80°C and 90°C is smaller than the difference between 90°C and 100°C since the radiation effect dominates in this aging condition.

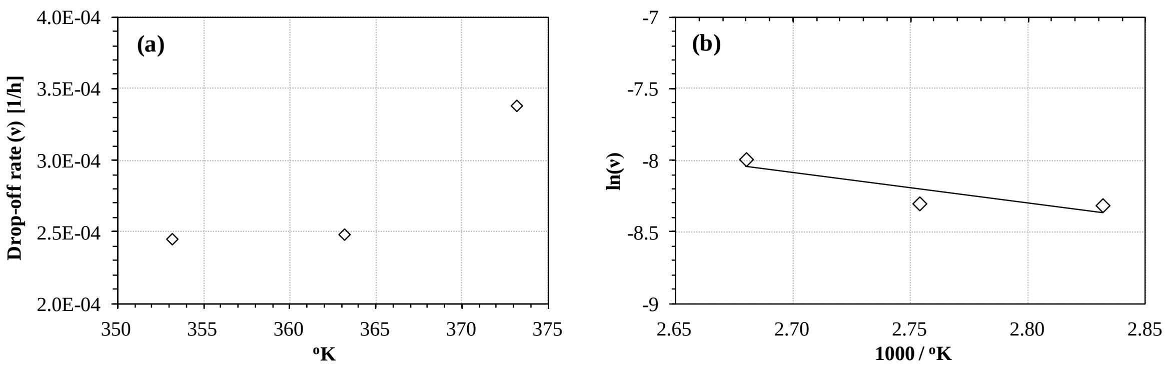

Figure 8. Drop-off rate (ν) vs. aging temperature (°K) at 100 Gy/h

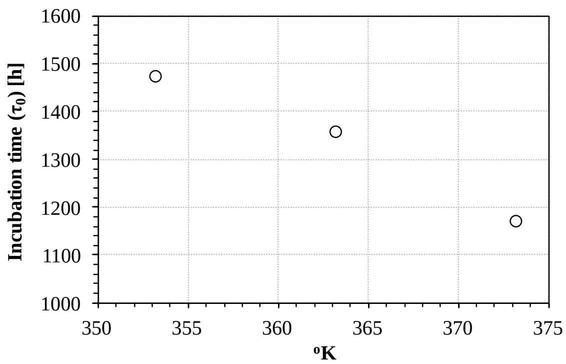

Figure 9. Incubation time (τ_0) vs. aging temperature (°K) at 100 Gy/h

It is worth noting that ν in this research is not equal to the degradation rate of one chemical reaction. The value of ν is determined from the trend of the EAB against time according to the Dichotomy Model. It is not calculated from the yield of a chemical reaction. There are complex reactions in the degradation of PVC. Minor cross-linking can improve the ductility but high cross-linking density decreases the EAB. On the other hand, rather than forming bonds, chain scission breaks the bonds between chains. There are at least these two reactions occurring simultaneously while unevenly contributing to the EAB of PVC. Therefore, ν determined from the EAB reflects the combined effects of myriad chemical reactions in the insulation. ν does not represent the degradation rate of a specific reaction.

5. CONCLUSION

PVC is one of the materials for cable insulation. The degradation of the insulation is a safety issue for nuclear power plants. The Dichotomy Model has been used to predict the degradation of the EAB of other insulation materials such as XLPE, EPR, and SIR. We applied the model to the EAB of SHPVC insulation exposed to heat and radiation, which are two major degradation factors in the field. It has been shown that the Dichotomy Model can pertinently represent the degradation of the EAB of SHPVC insulation.

There are two parameters in the model: τ_0 and ν. When the additives are sufficient, the EAB insignificantly changes as time elapses. This period of time is defined as τ_0. When the EAB curve exceeds τ_0, ν is used to represent the drop-off rate of the curve. This drop-off rate stands for the combined effects of myriad chemical reactions in the insulation. In other words, the proposed model is kinetics-based and the parameters are of physical significance.

The modeled results fit experimental data, which means the model is validated. In addition to the deterministic approach representing the value of the parameter for the model, Bayesian parameter estimation has also been used in this research developing a probabilistic approach to represent the parameter by a distribution denoting the uncertainty of drop-off rate. In conclusion, Dichotomy Model can be used to represent the reliability of PVC insulation exposed to heat and radiation.

References

[1] IAEA, "Assessment and management of ageing of major nuclear power plant components important to safety: In-containment instrumentation and control cables Volume I," International Atomic Energy Agency, Vienna, Austria IAEA-TECDOC-1188, December 2000.

[2] R. Duckworth and S. Davis, "Consequence of Activation Energy and Mechanical Properties in Harvested I&C Cables," presented at the ANS Winter Meeting and Nuclear Technology Expo, Washington, D.C., 2017.

[3] L. Fifield, "Tracking Aging in Nuclear Electrical Cable Polymers," presented at the ANS Winter Meeting and Nuclear Technology Expo, Washington, D.C., 2017.

[4] K. T. Gillen and R. L. Clough, "Time-temperature-dose rate superposition: a methodology for extrapolating accelerated radiation aging data to low dose rate conditions," *Polymer Degradation and Stability,* vol. 24, pp. 137-168, 1989.

[5] Y. Ohki, H. Kotani, D. Fujishima, and N. Hirai, "Similarity in Degradation Behavior by Heat and Irradiation Between Ethylene-propylene-diene Rubber and Silicone Rubber," presented at the IEEE Conference on Electrical Insulation and Dielectric Phenomenon, Fort Worth, TX, 2017.

[6] A. Shimada, M. Sugimoto, H. Kudoh, K. Tamura, and T. Seguchi, "Radiation ageing technique for cable life evaluation of nuclear power plant," *IEEE Transactions on Dielectrics and Electrical Insulation,* vol. 19, pp. 1768-1773, 2012.

[7] A. Shimada, M. Sugimoto, H. Kudoh, K. Tamura, and T. Seguchi, "Degradation distribution in insulation materials of cables by accelerated thermal and radiation ageing," *IEEE Transactions on Dielectrics and Electrical Insulation,* vol. 20, pp. 2107-2116, 2013.

[8] A. Shimada, M. Sugimoto, H. Kudoh, K. Tamura, and T. Seguchi, "Degradation mechanisms of silicone rubber (SiR) by accelerated ageing for cables of nuclear power plant," *IEEE Transactions on Dielectrics and Electrical Insulation,* vol. 21, pp. 16-23, 2014.

[9] T. Yamamoto and T. Minakawa, "The final report of the project of assessment of cable aging for nuclear power plants," Japan Nuclear Energy Safety Organization, Tokyo, Japan JNES-SS-0903, July 2009.

[10] IAEA, "Assessment and management of ageing of major nuclear power plant components important to safety: In-containment instrumentation and control cables Volume II," International Atomic Energy Agency, Vienna, Austria IAEA-TECDOC-1188, December 2000.

[11] S. Burnay, "A practical model for prediction of the lifetime of elastomeric seals in nuclear environments," presented at the ACS Symposium, Washington DC, 1991.

[12] K. T. Gillen and R. Bernstein, "Review of nuclear power plant safety cable aging studies with recommendations for improved approaches and for future work," Sandia National Laboratories, Albuquerque, New Mexico SAND 2010-7266, 2010.

[13] Y.-S. Chang and A. Mosleh, "Predictive model of the degradation of cable insulation subject to radiation and temperature," presented at the International Topical Meeting on Probabilistic Safety Assessment and Analysis (PSA), Pittsburgh, PA, 2017.

[14] Y.-S. Chang and A. Mosleh, "Physics-Based Model of the Degradation of Cable Insulation Subject to Radiation and Heat," presented at the IEEE Conference on Electrical Insulation and Dielectric Phenomenon, Fort Worth, TX, 2017.

[15] Y.-S. Chang and A. Mosleh, "Probabilistic Degradation Models for Cable Insulation in Nuclear Power Plants," presented at the ANS Winter Meeting and Nuclear Technology Expo, Washington, D.C., 2017.

[16] E. Rabiei, H. Chien, M. White, A. Mosleh, S. Iyer, and J. Woo, "Component Reliability Modeling Through the Use of Bayesian Networks and Applied Physics-based Models," in *Reliability and Maintainability Symposium (RAMS)*, Reno, Nevada, USA, 2018.

[17] E. Rabiei, E. L. Droguett, and M. Modarres, "A prognostics approach based on the evolution of damage precursors using dynamic Bayesian networks," *Advances in Mechanical Engineering,* vol. 8, p. 1687814016666747, 2016.

[18] E. Rabiei, E. L. Droguett, and M. Modarres, "Damage monitoring and prognostics in composites via dynamic Bayesian networks," in *Reliability and Maintainability Symposium (RAMS), 2017 Annual*, 2017, pp. 1-7.

[19] E. Rabiei, E. L. Droguett, and M. Modarres, "Fully Adaptive Particle Filtering Algorithm for Damage Diagnosis and Prognosis," *Entropy,* vol. 20, p. 100, 2018.

[20] E. Rabiei, E. L. Droguett, M. Modarres, and M. Amiri, "Damage precursor based structural health monitoring and damage prognosis framework," *Safety and Reliability of Complex Engineered Systems, vol,* pp. 2441-2449, 2015.

[21] H. A. Shnawa, "Thermal stabilization of polyvinyl chloride with traditional and naturally derived antioxidant and thermal stabilizer synthesized from tannins," *Journal of Thermal Analysis and Calorimetry,* vol. 129, pp. 789-799, 2017.

[22] L. Gervat and P. Morel, "Analysis of the main parameters affecting the change of color and physical properties of weathered PVC profiles," *Journal of Vinyl and Additive Technology,* vol. 2, pp. 37-43, 1996.

[23] C. S. Kong, G. H. Yoon, G. Khang, J. M. Rhee, and H. B. Lee, "Stabilization of nontoxic PVC formulation for gamma irradiation sterilization, II. Effect of antioxidants," *Bio-medical materials and engineering,* vol. 12, pp. 211-224, 2002.

[24] D. Turner, "Radiation crosslinking of rubber: Yields of hydrogen and crosslinks," *Polymer,* vol. 1, pp. 27-40, 1960.

[25] S. R. Scagliusi, E. L. Cardoso, and A. B. Lugao, "Effect of gamma radiation on chlorobutyl rubber vulcanized by three different crosslinking systems," *Radiation Physics and Chemistry,* vol. 81, pp. 1370-1373, 2012.

[26] H. Fang, J. Li, H. Chen, B. Liu, W. Huang, Y. Liu, *et al.*, "Radiation induced degradation of silica reinforced silicone foam: Experiments and modeling," *Mechanics of Materials,* vol. 105, pp. 148-156, 2017.

[27] F. Thominette, G. Metzger, B. Dalle, and J. Verdu, "Radiochemical ageing of poly (vinyl chloride) plasticized by didecylphthalate," *European polymer journal,* vol. 27, pp. 55-59, 1991.

[28] P. H. Daniels, "A brief overview of theories of PVC plasticization and methods used to evaluate PVC-plasticizer interaction," *Journal of vinyl and additive technology,* vol. 15, pp. 219-223, 2009.