

Issues with Approaches for Simulating Aging of Nuclear Power Plant Cable Materials Described in IEC and IAEA Documents

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ABSTRACT

Over the past 20 years, the IEC and the IAEA have published several Technical Documents describing recommended methods for carrying out accelerated radiation plus temperature aging of safety related cable materials in nuclear power plant environments. The methods described include the power law method, the time-dependent model, the DED approach and the Simplified Method approach. Because of the expected and observed changes in chemistry that occur as aging conditions transition radiation-temperature space, we highlight significant issues with the time-dependent and Simplified Method approaches. The DED approach and a recent modification (the MAC approach) can successfully handle the observed changes in chemistry for many important cable materials and therefore offer more confident accelerated simulations. Problems with the power law method are then briefly described. Also discussed are the significant issues that occur when trying to simulate the aging of semi-crystalline cable materials (e.g., EP, XLPE and XLPO) that show inverse-temperature effects. For these materials, degradation rates under radiation can increase as the aging temperature drops below ~60°C, in temperature regions where typical ambient aging conditions occur. A possible approach for dealing with such materials is suggested.

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INTRODUCTION

Over many decades, there has been a substantial effort devoted to developing reliable accelerated aging approaches for predicting polymer lifetimes when polymers are exposed to combined radiation plus temperature environments. Until recently, the two most applied approaches were referred to as the “Superposition of time-dependent data” developed by Burnay and co-workers^{1,2} and the “Superposition of DED (dose to equivalent damage) data” based on the time-temperature-dose rate superposition method developed by Gillen and Clough.^{3,4,5} These two approaches have been highlighted in the first and second edition of an IEC document^{6,7} as well as a 2000 IAEA document⁸ and a 2012 IAEA document.⁹ In addition, the 2012 IAEA document introduced a new approach, referred to as the “Simplified Method” based on work by Yamamoto and Minikawa.¹⁰ Also mentioned in all of these documents is the power law approach. More recently, a third IAEA document¹¹ came out suggesting combined environment accelerated aging conditions meant to prepare samples for simulating ambient conditions so the sample conditions could be tracked with various condition monitoring techniques.

The first goal of this paper is to show compelling evidence that the degradation chemistry differs across radiation-temperature (R - T) space based both on expectations and on experimental results that show that the shape of the degradation curves when plotted versus log of the aging time changes with position in (R , T) space. Since the time-dependent model explicitly assumes that the degradation chemistry remains unchanged for different combinations of radiation dose rate and temperature, this approach has important issues. Our next goal is to show that the DED approach can handle the observed changes in chemistry by discussing a recently described method¹² of utilizing this approach in a novel way by equally accelerating both the radiation initiation reactions and the thermally-initiated reactions. We refer to this approach as the MAC approach standing for Matched Accelerated Conditions. We will then show that the “Simplified Method” approach totally fails for reasons similar to the failure of the time-dependent method, based on the fact that the simulation condition leads to chemistry that is far different from the ambient aging conditions it is trying to simulate. We then note that the accelerated aging conditions recommended in the more recent 2017 IAEA document¹¹ results in degradation chemistry even more removed from ambient. We then briefly describe the problems inherent in the power law approach. Thus, of the four methods, only the DED approaches (original DED and new MAC modification) are of general utility. Finally we will note a significant issue (inverse-temperature effects^{13,14}) that occurs for many important EPR and XLPO materials and suggest a possible approach for accelerated aging simulations of these materials.

RESULTS AND DISCUSSION

CHANGING CHEMISTRY ACROSS R - T SPACE ARGUES AGAINST TIME-DEPENDENT MODEL

In a combined radiation-thermal environment, the initiation of the degradation chemistry comes from a combination of thermal initiation reactions and radiation initiation reactions. When radiation dose rates are lower and temperatures are higher, thermal initiation becomes more

important with the opposite true when temperatures are lower and dose rates are higher. The energy involved in radiation initiation (1.17 MeV and 1.33 MeV for Co^{60} for example) is much higher than thermal energies (0.025 eV at 20°C rising to only 0.036 eV at 150°C). This means that the thermal initiation reactions lead to free radicals at thermally weak regions of the polymer chain and/or its side chains. With radiation, the higher energy involved leads to a more random and diverse set of free radical species.¹⁵ Thus it is expected that the degradation chemistry will vary across R - T space as one moves from a radiation-dominated regime towards a more temperature dominated regime. We can expect to see this experimentally if the shapes of the degradation curves across R - T space change when plotted against log time or log dose and this is the case for a vast majority of the materials that have been examined to date. For instance, our results¹² for a chloroprene (CP) rubber and a chlorosulfonated polyethylene (CSPE) rubber are shown in Figure 1. For the CP material, the results at 46°C + 9180 Gy/h are close to R -dominated and the results at 115°C + 377 Gy/h are close to thermal-dominated. If the degradation chemistry was identical under the two aging conditions except for its acceleration, the shape of the curves versus log of aging dose would be identical and the shapes are clearly non-identical. Similar conclusions hold for the CSPE results.

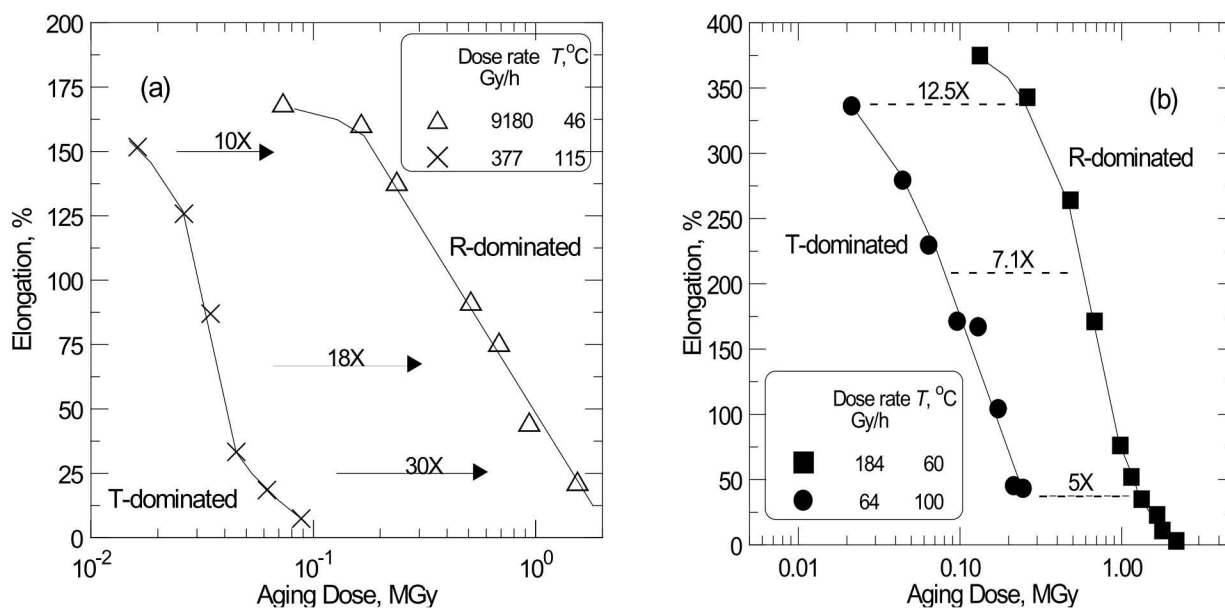


Figure 1. Tensile elongation results versus radiation dose. (a)- chloroprene rubber; (b)- CSPE rubber.

The JNES study¹⁰ examined several types of materials including EPRs, XLPEs, Silicone rubbers and PVCs. Even though their (R - T) studies only covered a fairly narrow temperature range (80°C to 100°C for the EPRs, XLPEs and PVCs and 100°C to 135°C for the silicone rubbers), the expected evidence for changes in chemistry were clearly present as seen in Figure 2 for an EPR and an XLPE material and Figure 3 for a silicone rubber and a PVC material. It is therefore clear that the expected changes in chemistry from differences in initiation reactions for radiation versus thermal initiation are experimentally confirmed for many types of materials, leading to non-identical shapes of the degradation curves versus position in R - T space. This conclusion is

in direct conflict with the time-dependent model which notes in all references that the model predicts the same degradation chemistry across R - T space and therefore that the curve shapes should be identical when plotted versus log of dose or log of time.

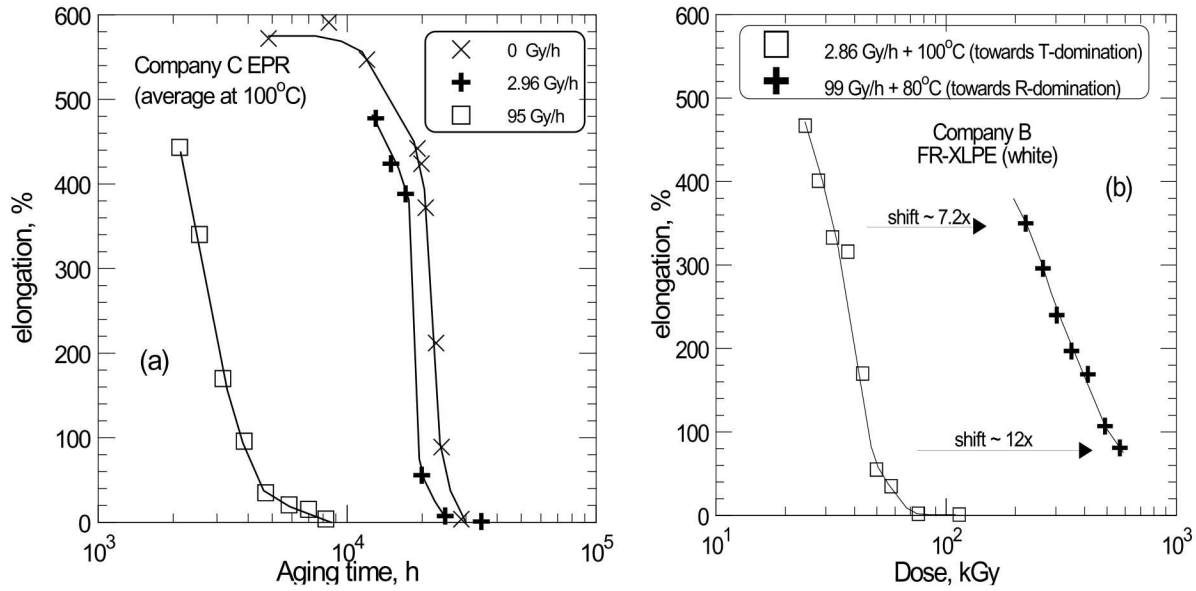


Figure 2. Tensile elongation results versus radiation dose. (a)- EPR from Company C; (b)- FR-XLPE white insulation from Company B.

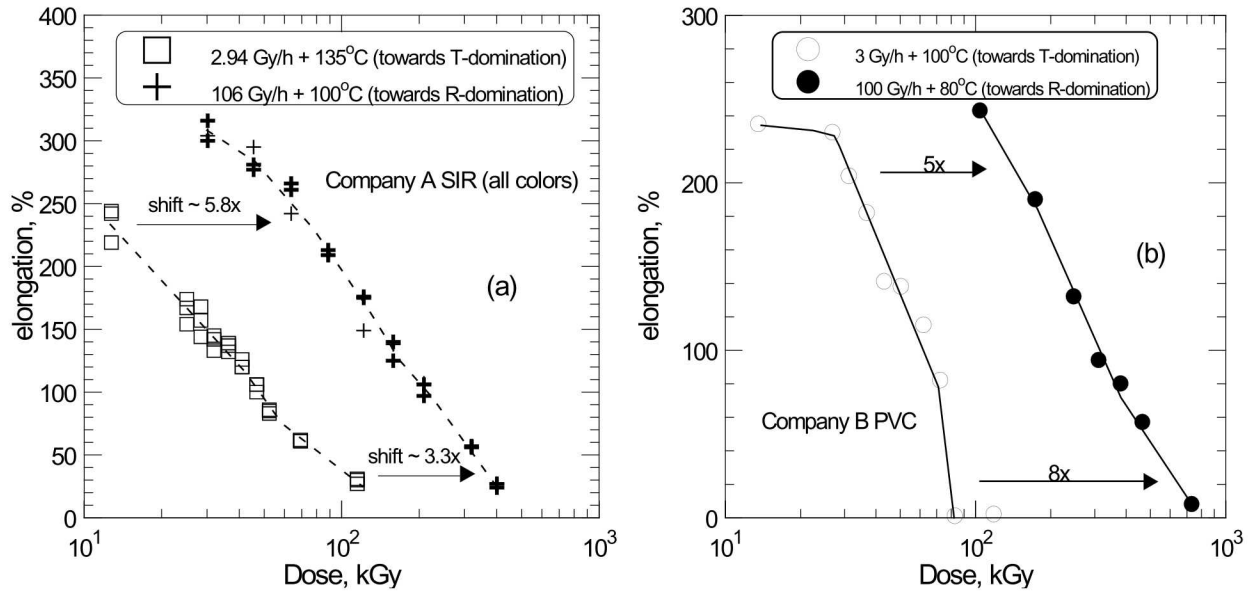


Figure 3. Tensile elongation results versus radiation dose. (a)- SiR from Company B; (b)- PVC from Company B.

THE MAC APPROACH CAN HANDLE CHEMISTRY CHANGES

The Matched Acceleration Condition (MAC) approach¹² is a recently updated version of the “Superposition of DED (dose to equivalent damage) data” approach where the latter is based on the time-temperature-dose rate (t - T - R) superposition method.^{3,4,5} The (t - T - R) superposition method assumes that if the temperature degradation rate is increased by a factor x (based on the Arrhenius activation energy from thermal-only aging) and at the same time, the radiation initiation rate is also increased by the same factor x (through an increase in dose rate by this same factor x), the resulting combined environment degradation rate will increase by the same factor x . This approach has traditionally been applied by selecting a failure criterion for the degradation variable of interest and determining the dose required to reach this constant degradation value versus the combined environment aging conditions. For example, we conducted an extensive study at 22 different combinations of radiation and temperature on a CR cable jacket.⁴ Figure 4a shows the radiation dose required to reach 50% elongation (Dose to Equivalent Damage or DED) versus dose rate and temperature. To use t - T - R superposition to make predictions at 50°C versus dose rate, the model requires the thermal activation energy E_a which is 89 kJ/mol above 70°C and 71 kJ/mol below this temperature.¹⁶ The next step is to horizontally shift each data point from their temperature and dose rate conditions to the temperature of interest assuming the model is valid. For instance, to shift the data point at 74°C + 112 Gy/h to 50°C, the E_a values imply a shift factor of 6.67. The model then assumes that in going to 50°C, the dose rate will drop by this same factor of 6.67 or from 112 Gy/h to 16.8 Gy/h as shown by its 6.67x drop to the filled in square on Figure 4a. Similar procedures are used to shift all of the 22 data points to 50°C with the results at 50°C shown on Figure 4b. The excellent superposition for this material and many other materials^{3,4} provides evidence for the assumptions underlying the t - T - R superposition model.

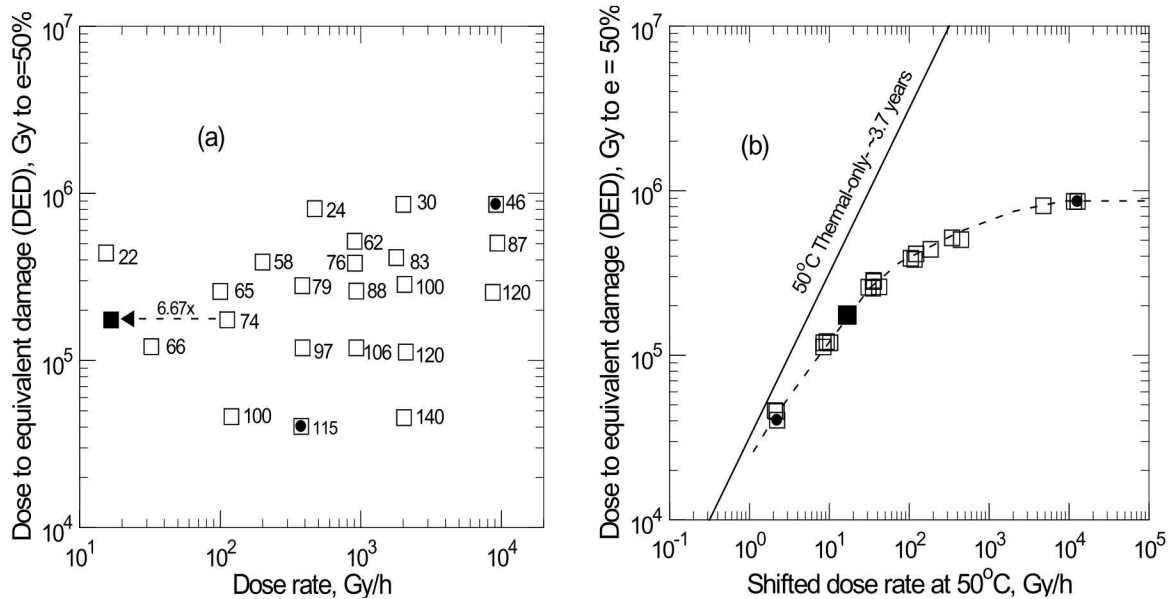


Figure 4. (a)- Dose required to reach 50% elongation versus dose rate and temperature in °C (the numbers next to the data points). (b)- t - T - R superposed results from (a) at 50 °C. The most T -dominated and most R -dominated have solid circles inside the squares (see Figure 1a).

Notice that the predicted results at 50°C seem to level out at high dose rates where radiation initiation dominates and the results at low dose rates approach the thermal-only result predicted at 50°C⁵ which is a constant time line of 3.7 years. Since typical worst-case ambient aging conditions for safety equipment like cables in nuclear power plants are often around 50°C + 0.1 Gy/h, these results predict that thermal-only effects will dominate for this chloroprene material and 50% elongation will be reached in ~ 3.7 years. It is also worth noting that the results plotted in Figure 1a (46°C + 9180 Gy/h and 115°C + 377 Gy/h) represent the data points closest to *R*-domination and *T*-domination, respectively and again reflect the expected changes in degradation chemistry.

The *t-T-R* superposition approach typically examines degradation to a given level (often based on a select failure criterion), which eliminates examining much of the dose-dependent data. Our recent improvement (the MAC approach) considers the entire dose dependent degradation curve.¹² The basis for the MAC approach can be seen by plotting the 22 aging conditions on a plot of log dose rate versus inverse absolute temperature as shown in Figure 5. As is clear from the comparison of Figure 5 with Figure 4 and as indicated on Figure 5, the experiments done at low temperatures and higher dose rates are closer to radiation domination and the experiments done at higher temperatures and lower dose rates are approaching thermal-domination. All of the MAC lines shown have Arrhenius slopes consistent with the Arrhenius behavior of this material¹⁶ where the activation energy is 89 kJ/mol above 70°C and 71 kJ/mol below 70°C. Each line is placed horizontally such that it approximately intersects several of the aging conditions which were planned to be approximately consistent with a MAC line. Thus the aging conditions along a MAC line would be consistent with the equal acceleration assumptions underlying the *t-T-R* superposition model, implying that experimental data along a MAC line should superpose when plotted versus radiation dose if the assumptions underlying *t-T-R* superposition are valid.

Plots of normalized elongation versus radiation dose along the 6 MAC lines of Figure 5 are shown on Figure 6. The excellent superposition along all MAC lines is consistent with the assumptions of the model even for very large acceleration factors (~120 for MAC-3 and ~87 for MAC-4). Although the usual changes in degradation chemistry are observed across *R-T* space (Figure 1a), the MAC approach gives us a way to handle such changes and make valid predictions for a given ambient condition by planning experiments along a MAC line that intersects the ambient conditions and therefore properly accelerates the responsible degradation chemistry. In our previous publication¹² the MAC approach was shown to be consistent with results for other materials including an EPR and a CSPE.

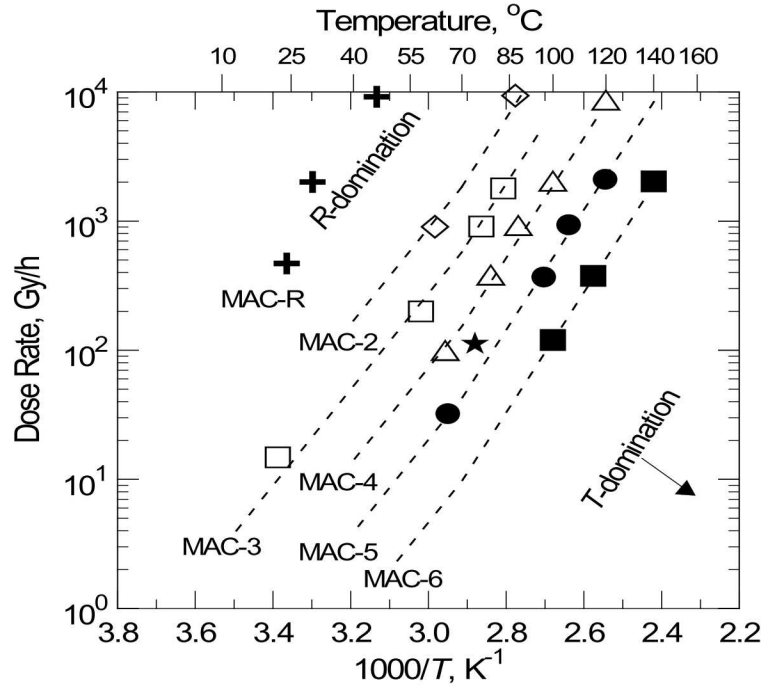


Figure 5. The 22 aging conditions for the chloroprene rubber. The dashed lines give lines indicative of Matched Accelerated Conditions (MAC lines).

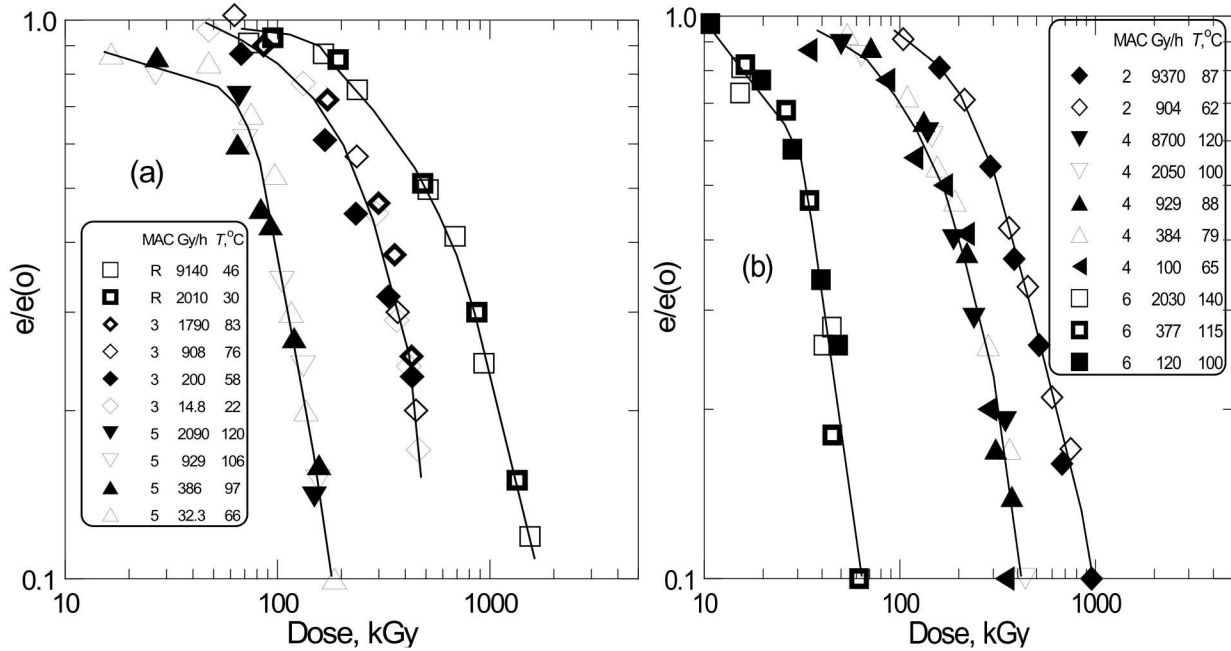


Figure 6. (a)- $e/e(0)$ versus radiation dose for aging experiments close to the MAC-R, MAC-3 and MAC-5 lines of Figure 5. (b)- $e/e(0)$ versus radiation dose for aging experiments close to the MAC-2, MAC-4 and MAC-6 lines of Figure 5.

PROBLEMS WITH THE SIMPLIFIED METHOD APPROACH

The Simplified Method approach for simulating combined radiation plus thermal ambient aging conditions was developed by JNES,¹⁰ it involves a six step procedure and the method was highlighted in the 2012 IAEA document.⁹ The JNES approach for their EPR insulation from Company C is shown on Figure 7. The first step involves using the DED superposition approach to select “Temporary EQ conditions” at 100°C that are equivalent, according to the DED model, to the ambient aging conditions (in this case 60 years at 50°C + 0.1 Gy/h or a total dose of 52.6 kGy). These ambient conditions corresponds to the open star on Figure 7. Assuming a thermal-only E_a of 15 kcal/mol (62.8 kJ/mol), leads to a DED equivalent of 2.62 yr at 100°C + 2.29 Gy/h (the closed star in the figure). Their accelerated data is generated at 9 aging conditions- 80°C, 90°C and 100°C at 3, 18 and 100 Gy/h (plotted as open circles, squares and diamonds), They then use an iterative approach to find the DED elongation (123% for the present case) that when shifted to 100°C for the 80°C and 90°C results (filled circles and squares- 100°C data requires no shifting) gives the overall best superposition curve for the shifted results. The 100°C shifted curve at 100 Gy/h gives the accelerated aging conditions for the Simplified Method of 100°C + 100 Gy/h for 3299 h.

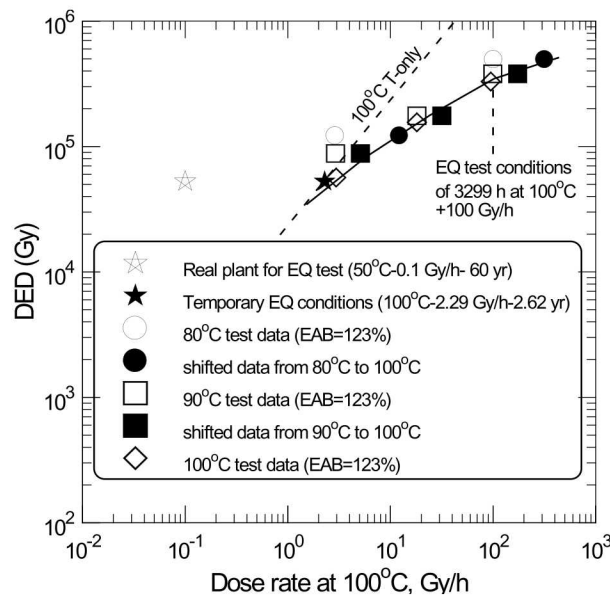


Figure 7. JNES Simplified Method approach applied to their EPR from Company C¹⁰.

To indicate the problems with the Simplified Method approach, we plot the ambient condition (50°C + 0.1 Gy/h) and the accelerated aging condition for 60 years resulting from the 6 step analysis (3299 h at 100°C + 100 Gy/h) on a MAC-type plot of log of the dose rate versus inverse absolute temperature with the results shown on Figure 8. As indicated on the Figure, the Simplified Method accelerates the time by a factor $a(t) = 159$, the radiation by $a(R) = 1000$ and the temperature by $a(T) = 22.9$, so the radiation undergoes much greater acceleration than temperature pushing the degradation chemistry much closer to radiation domination [$a(R)/a(T) = 43.7$]. Therefore the Simplified Method simulation conditions would be expected to not simulate the chemistry expected under ambient conditions. Thus subsequent use of the aged samples for

condition monitoring information or for subsequent Loss of Coolant Accident simulations would be of questionable value.

If this material is a well-behaved material (e.g., not a material showing inverse-temperature behavior as will be discussed below), the proper acceleration approach would involve applying the MAC approach. Using the assumed E_a of 62.8 kJ/mol used by JNES together with the MAC approach, the calculations leading to three such proper simulations of the degradation chemistry occurring under ambient conditions would be 2.62 years at 100°C + 2.29 Gy/h, 1.55 years at 110°C + 3.88 Gy/h and 0.94 years at 120°C + 6.4 Gy/h as shown on the Figure. It is interesting to note that the first step in the Simplified Method approach follows the MAC approach (2.62 years at 100°C + 2.29 Gy/h). As indicated on the Figure, the MAC simulations equally accelerate the aging time and the initiation rates for radiation and thermal, e.g., [$a(T) = a(R) = a(t)$, so the ratio of $a(R)$ over $a(T)$ is unity]. This strongly suggests that the chemistry under accelerated conditions would reflect the chemistry expected under ambient, again assuming that this material does not exhibit inverse-temperature behavior. This also implies that any condition monitoring method used on the accelerated samples should successfully reflect the chemistry expected under ambient conditions.

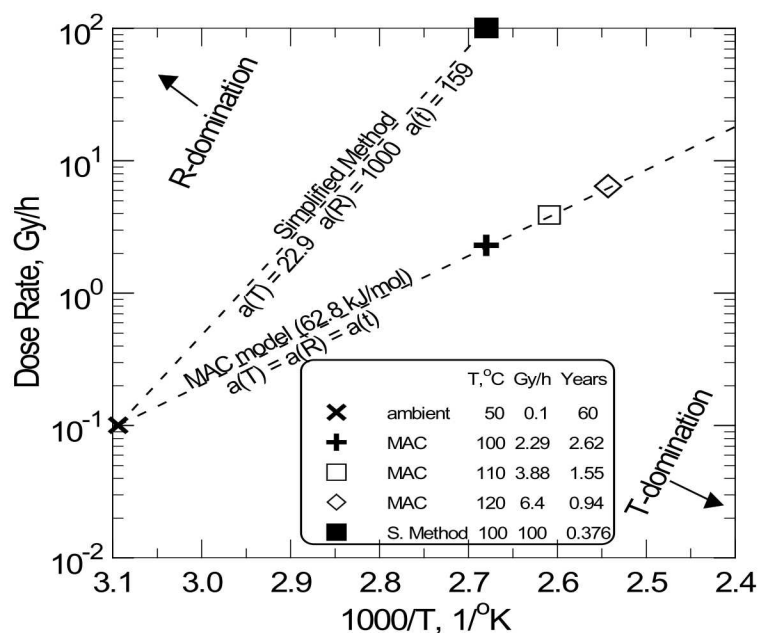


Figure 8. A MAC style plot showing the Simplified Method conditions used by JNES (0.376 years at 100°C + 100 Gy/h) to stimulate ambient aging conditions (50°C + 0.1 Gy/h for 60 years) together with conditions appropriate to the MAC approach.

As indicated in Figure 8, proper MAC modeling at 100°C involves 2.29 Gy/h. Since JNES ran an experiment close to these conditions at 2.96 Gy/h + 100°C (Figure 2a), we can compare these results to their 100°C thermal-only results plus their conditions run close to those specified from the Simplified Method approach (100°C + 95 Gy/h), data also shown in Figure 2a. This figure indicates that the results at 2.96 Gy/h (near proper accelerated aging conditions of 2.29 Gy/h) are close to the thermal-only results (2.29 Gy/h would be even closer). The shape of the results at 95

Gy/h (close to the Simplified Method condition of 100 Gy/h) differs substantially, confirming differences in degradation chemistry. Additional evidence of differences in chemistry comes from plots of elongation against tensile strength where changes in chemistry are apparent as one moves from T -dominated towards R -dominated, even though the 80°C + 102 Gy/h has not reached radiation domination (Figure 9).

Table I gives a summary of the 14 materials studied in the JNES Final Report¹⁰ showing the plant conditions simulated, the assumed E_a values and the values of $a(R)$, $a(T)$, and $a(R)/a(T)$ appropriate to the Simplified Method aging conditions (always done at 100°C + 100 Gy/h). As indicated in the table, the $a(R)/a(T)$ values ranged from ~21 to ~127, again indicative of simulated aging closer to the radiation limit and therefore generating chemistry not matching ambient aging conditions.

In order to simplify their 6 step procedure for choosing accelerating conditions based on the Simplified Method, JNES derived the following equation (eq. A-3 in the 2012 IAEA Report⁹) for selecting the accelerated conditions, where a is the acceleration factor, E_a is the thermal activation energy, T_1 , D_1 and t_1 are the ambient aging conditions (temperature, dose rate and time) and T_2 , D_2 and t_2 are the accelerated simulation conditions.

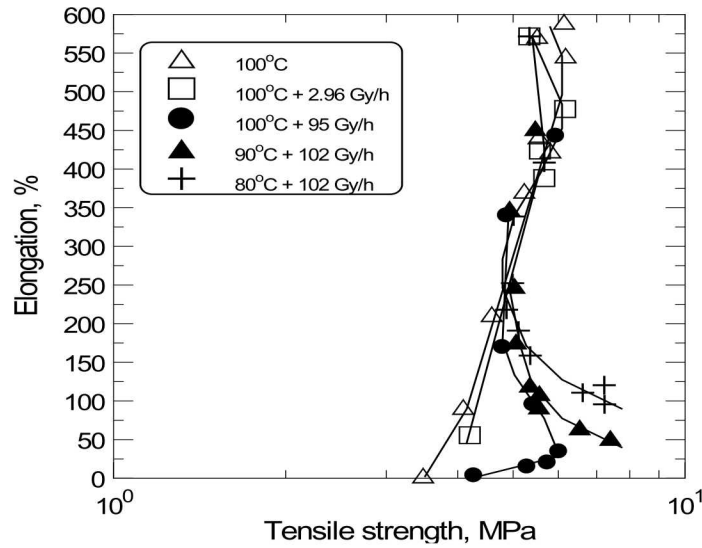


Figure 9. Elongation versus tensile strength for EPR from Company C.

Table I. Summary of materials studied by JNES.¹⁰

Cable material	Plant conditions	Assumed E_a	Aging t , h	$a(R)$	$a(T)$	$a(t)$	$a(R)/a(T)$
XLPE, Company A	60 yr/40C/0.03 Gy/h	15 kcal/mol	706	3333	48	745	69
XLPE, Company B	60 yr/40C/0.03 Gy/h	15 kcal/mol	1076	3333	48	489	69
XLPE, Company C	60 yr/50C/0.2 Gy/h	15 kcal/mol	4129	500	23	127	21.8
FR-XLPE, Company A	60 yr/40C/0.1 Gy/h	15 kcal/mol	1798	1000	48	292	20.7
FR-XLPE, Company B red	60 yr/40C/0.1 Gy/h	15 kcal/mol	2241	1000	48	235	20.7
EPR, Company C	60 yr/50C/0.1 Gy/h	15 kcal/mol	3299	1000	23	159	43.7
FR-EPR, Company A red	15 yr/40C/0.3 Gy/h	15 kcal/mol	1429	333	48	368	6.9
FR-EPR, Company B white	50 yr/50C/0.1 Gy/h	15 kcal/mol	4272	1000	23	123	43.7
FR-EPR, Company C white	50 yr/50C/0.1 Gy/h	15 kcal/mol	3555	1000	23	148	43.7
SiR, Company A	60 yr/50C/0.15 Gy/h	11.3 kcal/mol	2100	667	11	250	62.9
SiR, Company B	60 yr/50C/0.1 Gy/h	9.9 kcal/mol	1493	1000	7.9	352	127
SiR, Company C	60 yr/50C/0.1 Gy/h	12 kcal/mol	1413	1000	12	372	82
SHPVC, Company A black	60 yr/40C/0.05 Gy/h	15 kcal/mol	984	2000	48	534	41.5
SHPVC, Company B	60 yr/40C/0.05 Gy/h	15 kcal/mol	957	2000	48	550	41.5

$$a = \frac{t_1}{t_2} = \left[e^{\frac{E_a}{R} \left(\frac{1}{T_1} - \frac{1}{T_2} \right)} * \frac{D_1}{D_2} \right]^b * \frac{D_2}{D_1}$$

The constant b is dependent upon insulation type (0.65 for FR-EPDM, 0.5 for XLPE, FR-XLPE and EPDM, 0.35 for SHPVC and 0.25 for SIR). The values of b were derived based on analysis involving the 6 step Simplified Method. For the example given in the IAEA Report⁹ (acceleration $a = 107.2$), the thermal acceleration (50°C to 100°C with E_a of 15 kcal/mol) is a factor of 22.9, whereas the dose rate acceleration (0.2 to 100 Gy/h) is a factor of 500, leading to issues similar to those discussed earlier (Figure 8). Since the acceleration a in the equation is a constant, time-independent value when comparing two aging conditions, the equation implies that the acceleration between any two aging conditions will be constant versus degradation (unchanging chemistry). As seen earlier, this is typically in conflict with experimental results with the exception of data obtained under Matched Accelerated Conditions.¹² Therefore, we conclude that the equation is of little value.

Even more problematic than the choice of carrying out simulated aging at 100°C + 100 Gy/h, the more recent IAEA document¹¹ seems to suggest combined environment aging at 50°C + 500 Gy/h to create aged materials for correlations with condition monitoring methods. For typical ambient conditions of 50°C + 0.1 Gy/h, this corresponds to $a(R) = 5000$ and $a(T) = 1$ and therefore chemistry so different from ambient that the results would typically be of little value. Looking back, for instance, at the t - T - R superposed results at 50°C for chloroprene (Figure 4b), it is clear that 50°C + 500 Gy/h is close to the radiation-dominated region and very far removed from typical temperature-dominated ambient conditions of 50°C + 0.1 Gy/h.

For the materials studied by JNES shown in Table I, it should also be pointed out that 9 of the 14 materials are XLPEs and EPRs. For these materials, inverse temperature effects are often observed below ~60°C where degradation rates at a constant dose rate become faster as the aging temperature drops.^{13,14} Since ambient aging conditions are usually lower than 60°C and the

JNES studies only looked at (*R-T*) aging at 80°C, 90°C and 100°C, the inverse temperature regions of interest to ambient aging conditions were not examined when deriving accelerated conditions using the 6-step procedure. Later in this paper we look more carefully at the complications caused by inverse temperature effects. It should be noted that if inverse-temperature effects are relevant to the EPR from Company C, the MAC accelerations shown in Figure 8 would be inappropriate.

PROBLEMS WITH THE POWER LAW APPROACH

As noted in International documents⁹ describing the power law extrapolation model, it is based on the extrapolation of radiation aging data obtained under isothermal conditions at several dose rates and is generally limited to temperatures near ambient (i.e., <40°C). The DED is assumed to be proportional to the dose rate to the power n , where n typically ranging from 0 (no dose rate effect) to 0.3. The main basis for this approach comes from a 1987 publication by Wilski.¹⁷ Unfortunately there are two major problems with Wilski's analyses. First of all, it was assumed that the data analyzed under "ambient temperature" conditions represented constant aging temperature versus dose rate. However, it turns out that typical exposures to Co-60 radiation results in the ambient temperature increasing as the dose rate increases due to the radiation energies involved. For instance, "ambient aging data" were used for several materials from our earliest paper on radiation aging¹⁸ when the actual "ambient temperatures" varied from 22°C at low dose rate to 38°C at the highest dose rate. A second problem had to do with the presence of diffusion-limited oxidation (DLO) effects at the highest dose rates for three of our materials (PVC, chloroprene rubber and chlorosulfonated polyethylene) that he analyzed. We suspect that similar problems existed for many of the other "constant temperature" materials he looked at for similar reasons. It is now generally accepted that the shapes of most DED curves versus dose rate for air-aging at a constant temperature resemble curves II or III in Figure 10, a figure that we introduced in 1991⁵ and has been used in several International documents starting in 1996.⁶

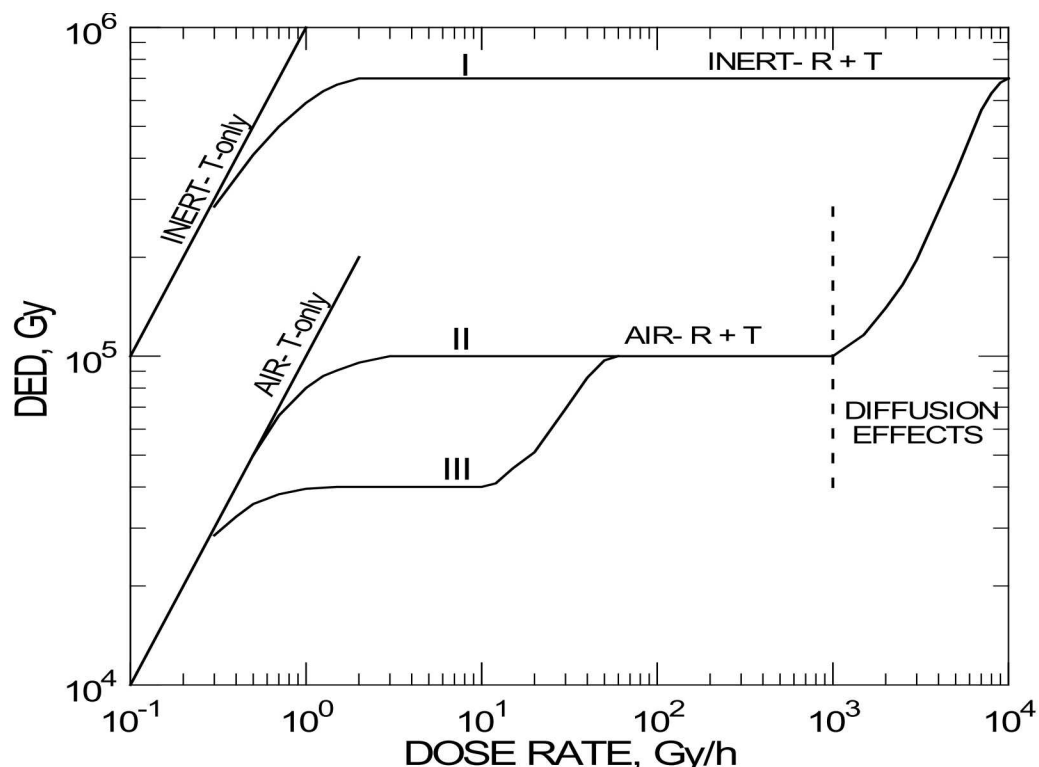


Figure 10. Generic representation of expected dose rate effect behavior at a constant aging temperature for inert aging (type I behavior) and for air aging (types II and III behaviors).

For air aging where data is shifted to a constant aging temperature using the DED approach, many materials have been shown to follow a type II curve shape,^{4,5,12} including, for instance, the chloroprene rubber results at 50°C shown in Figure 4(b). For such materials, dose rate effects disappear at high enough dose rates where the aging temperature has little effect with a smooth transition to the thermal-only line at low dose rates where the temperature dominates degradation. Type III behavior was found for a PVC material^{3,4,5} caused by the thermal breakdown of ROOH species created by radiation initiation as seen in Figure 11.

The same materials (PVC, chloroprene, chlorosulfonated polyethylene) of ours that Wilski concluded showed power law behavior instead displayed Type II and Type III shapes when properly analyzed using the DED approach taking account of DLO effects and non-constant “ambient” aging temperatures. Since it is clear that Type II and Type III behaviors are inconsistent with a power law assumption, we conclude that the power law approach is typically of little utility. The power law approach also suffers from the fact that, at the constant temperature being probed, the degradation chemistry changes as the dose rate goes from high dose rates where radiation initiation dominates to low dose rates where temperature effects enter.

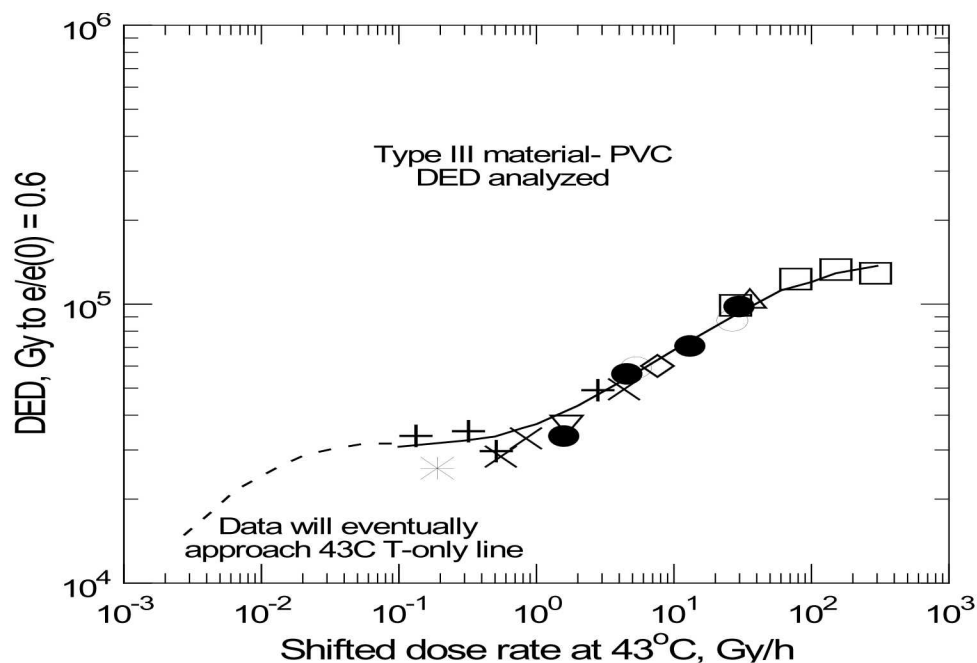


Figure 11. Results for a Type III material (PVC) analyzed at 43 °C using the DED approach.

INVERSE TEMPERATURE COMPLICATIONS

So-called inverse-temperature effects, has been documented for several EPR/EPDM and XLPE/XLPO materials.^{13,14} For these materials, drops in temperature below ~60°C at a constant dose rate results in faster degradation. Application of the DED superposition approach gives reasonable superposition for temperatures at ~60°C and above but totally fails to give superposition for data taken at lower temperatures. Typical DED results^{13,14,19} are shown for a Brandrex XLPO cable insulation in Figure 12a. It is obvious that the results below 60°C (filled squares for experiments run at 22°C and 41°C) show inverse temperature behavior since they reach 100% elongation quicker than samples aged at higher temperatures. DED superposition at 60°C using the appropriate E_a of 72 kJ/mol¹⁹ leads to the results shown in Figure 12b, where the results at 60°C and above give nice superposition. To be consistent with time-temperature-dose rate superposition and MAC assumptions, the 22°C and 41°C results would be expected to give results indicated by the four Xs shown in Figure 12a.

Since the JNES study¹⁰ of their 9 EPR and XLPE materials only looked at combined environment aging at temperatures ranging from 80°C to 100°C, analyses of their data completely misses the potential complications caused by inverse temperature effects. Since they were attempting to simulate aging conditions at 40°C and 50°C for each of the 9 materials, the possible presence of inverse-temperature effects would imply even greater chemistry disparities between accelerations using the Simplified Method approach and the chemistry operative under ambient conditions. As an example, we show in Figure 13 elongation data taken at 41°C + 69 Gy/h for the Brandrex XLPO material.¹³

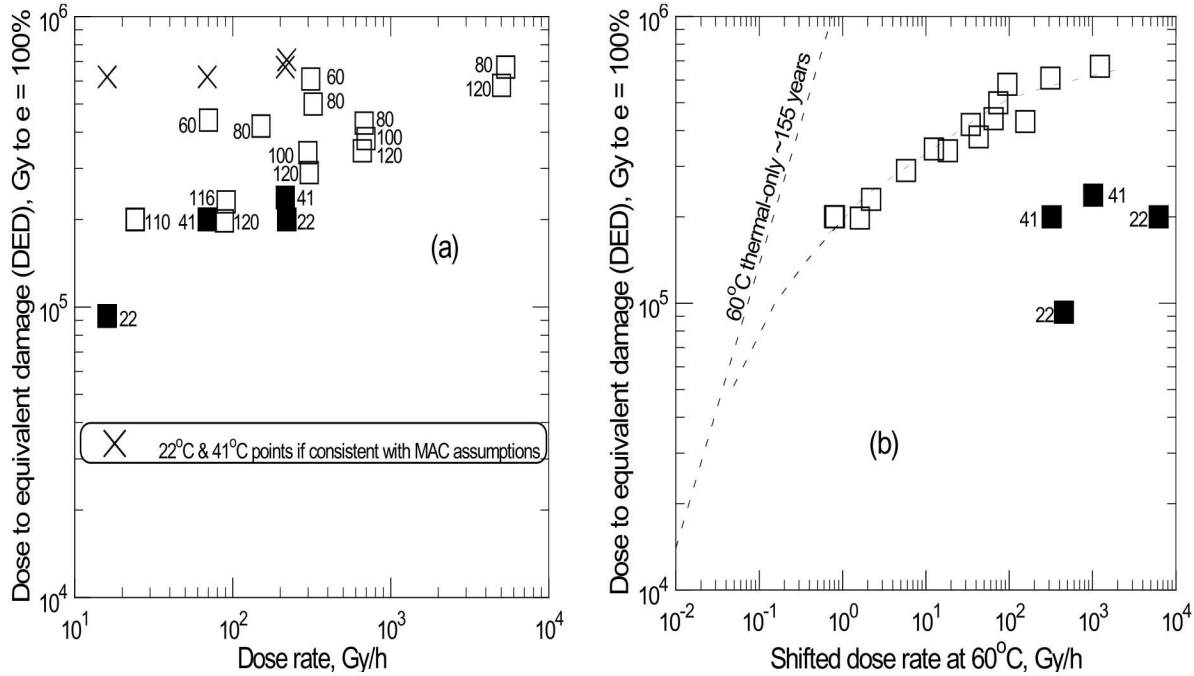


Figure 12. (a)- DED results to $e = 100\%$ versus dose rate and temperature (numbers by data points in °C). (b)- t - T - R superposed results at 60°C using $72 \text{ kJ/mol } E_a$ value.

If we ignore inverse-temperature effects and assume that the t - T - R superposition approach is valid, using the measured E_a of 72 kJ/mol ,¹⁹ equivalent accelerated aging conditions at 60°C would be 60°C + 332 Gy/h. Although we do not have data under these exact conditions, we do have data at 60°C + 317 Gy/h¹³ which is a reasonable approximation. The elongation results for these two aging conditions are shown in Figure 13. As seen in the figure, if we were attempting to simulate 1 year of aging at 41°C + 69 Gy/h (605 kGy dose) using accelerated conditions of 60°C + 317 Gy/h, we would predict that the elongation dropped to ~100%. This would totally underestimate the actual degradation at 41°C + 69 Gy/h (totally dead after ~300 kGy) caused by inverse-temperature effects. In other words, the expected chemistry in the inverse-temperature region can be vastly different from that found at higher temperatures. This is seen dramatically from data showing the correlation of elongation results with uptake factor and percentage gel measurements^{13,14} (Figure 14). Since the uptake factor decreases and the percentage gel increases with aging at combined environment conditions where the temperature is 60°C or higher, but the opposite occurs in the inverse-temperature region, it is clear that there is a dramatic switch in the degradation chemistry from crosslinking processes dominating scission

processes at higher temperatures to the opposite in the inverse temperature region. Since combined environment aging temperatures used in the JNES study for such potentially inverse-temperature materials were at 80°C and higher, this dramatic change in chemistry expected under ambient conditions for potentially inverse-temperature materials was not accounted for. In addition, given the huge differences in chemistry found in the inverse temperature region and the fact that ambient aging temperatures are in such regions, any condition monitoring assessments must clearly be done on samples aged at or near ambient temperatures to ~100%.

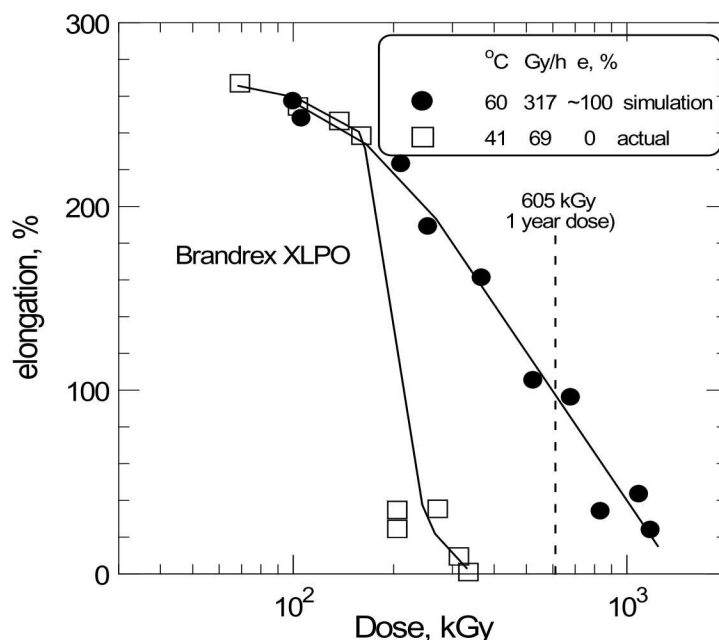


Figure 13. Brandrex XLPO data. Elongation at the conditions indicated.

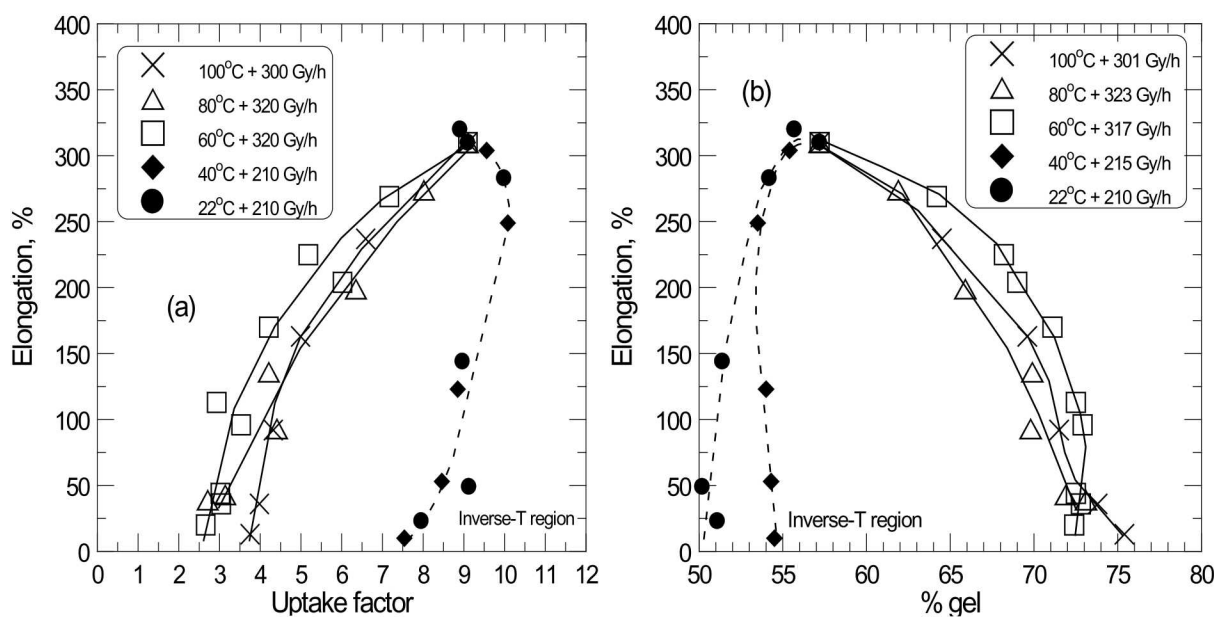


Figure 14. Brandrex XLPO. (a)- elongation versus uptake factor. (b)- elongation versus percentage gel.

POSSIBLE APPROACH FOR INVERSE TEMPERATURE MATERIALS

It should now be clear that simulation of typical nuclear power plant ambient aging conditions using higher than ambient temperature accelerated conditions will not be viable for materials showing inverse temperature behavior. This means that any material of interest must first be screened to determine whether inverse temperature behavior exists before accelerated aging studies are attempted. If inverse temperature effects are not present, the MAC approach is clearly the acceleration method of choice, an approach that requires sufficient thermal-only aging experiments so that reasonable estimates of the appropriate Arrhenius activation energies are available. If, on the other hand, inverse temperature effects are present, it is clear that studies must be done versus dose rate at the actual ambient temperature of interest since higher temperature radiation exposures can slow down the degradation rate and lead to chemistry that differs from ambient aging conditions. In other words, the two viable higher temperature accelerated aging simulation approaches (DED superposition and the MAC approach) cannot be confidently used to simulate ambient aging conditions for materials that show inverse temperature behavior. Thus it is critically important to conduct some combined environment exposures at temperatures near ambient to determine whether a given material is an inverse temperature material. If inverse temperature behavior is confirmed, it is interesting to note that, contrary to normal behavior, inverse temperature materials age faster than at higher temperatures (see Figure 12a). This implies that combined environment experiments at or near ambient aging temperatures are viable down to lower dose rates than would normally be expected in reasonable time frames, making such experimental conditions more viable.

It turns out that remarkably important studies were reported on many years ago by Reynolds and co-workers²⁰ at the University of Virginia. They exposed four different commercial EPDM/EPM insulations and two different commercial XLPE insulations used in nuclear power plant safety cables to aging at room temperature ($\sim 22^{\circ}\text{C}$) + five different radiation dose rates ranging from ~ 4 Gy/h to ~ 2500 Gy/h with aging times approaching 5 years at the 4 Gy/h dose rate. For elongation measurements of every one of these materials, they concluded that dose rate effects were absent from ~ 4 Gy/h to at least 40 Gy/h. Two of these materials were Brandrex XLPO (their material labeled XLPE1) and Okonite EPR (their material labeled EPDM4), materials found to be inverse temperature materials by Sandia^{13,14,21}.

For averages of three insulation colors (red, white and gray) of the Brandrex XLPO, Figure 15a shows their room temperature results for the dose required for the elongation to drop to 70% and 30% of its original value versus dose rate. In addition, we estimated from their Figure 4 the doses required to reach 90% of initial which are also plotted on the figure. The lack of dose rate effects at low dose rates is clear and their conclusion that DLO effects likely influence their data at the two higher dose rates for their 0.98 mm thick samples is consistent with calculations.²² Sandia's study of the same material (Brandrex XLPO) at 22°C gave very similar results with $e/e(0)$ reaching 0.3 after 9.5×10^4 Gy at $22^{\circ}\text{C} + 16.8$ Gy/h and 2×10^5 Gy at $22^{\circ}\text{C} + 210$ Gy/h. Using Sandia's results for thermal-only aging (no such results were reported by the Reynolds group), the constant time-lines appropriate to $e/e(0) = 0.9$ and 0.3 are also shown on the figure. The horizontal (no dose rate effect) behaviors must eventually transition to the constant time-lines.

Although the exact shape of these transitions is unknown, the dashed lines shown on the figure show what might be generally expected based on results from numerous past materials.^{4,19,21,22} Since 0.1 Gy/h is often noted as the worst-case expected ambient dose rate, 15, 30 and 45 year doses at this dose rate are noted on the figure. These results give initial strong confidence that the Brandrex XLPO insulation material should be viable for 60 years at 22°C + 0.1 Gy/h.

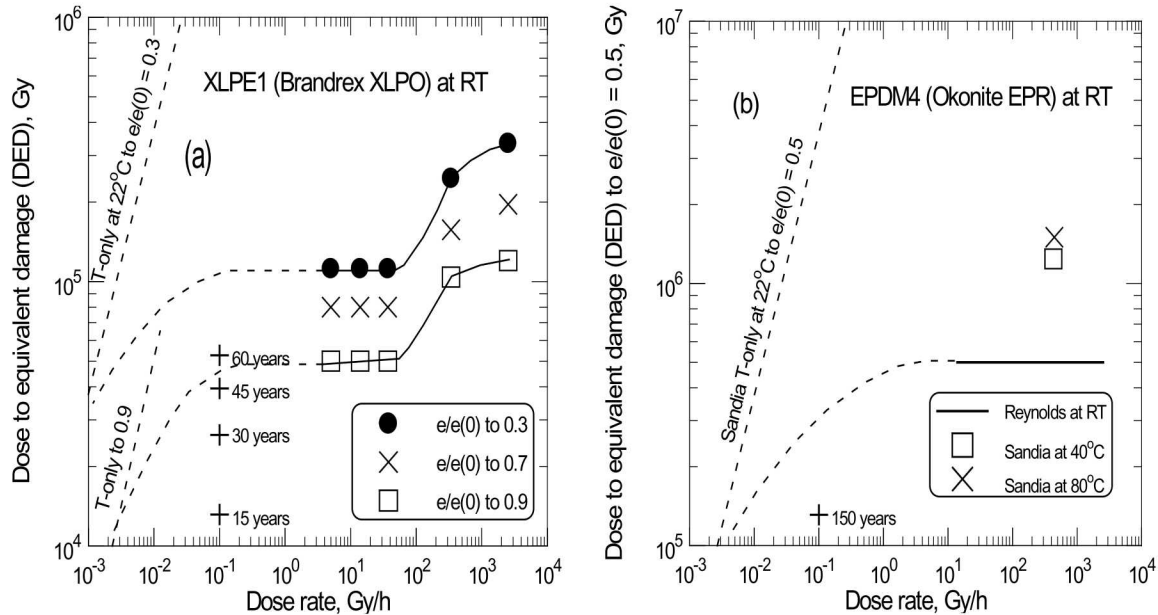


Figure 15. (a)- Room temperature elongation results for Brandrex XLPO. (b)- Room temperature elongation results for Okonite EPR plus Sandia results at 40°C and 80°C

Further confirmation of this conclusion could involve removing a small piece of cabling after 15, 30 and 45 years to confirm that little damage has occurred to the insulation. In addition, the removal of such ambient-aged samples would allow confident condition monitoring measurements (e.g., % gel, uptake factor, etc.) to be made on samples reflecting the chemistry relevant to ambient aging.

Based on the low temperature radiation results for the five EPM /EPDM samples studied by Reynolds, et. al.²⁰, it appears that these materials generally age even slower than the XLPE materials. For all of their EPR cable materials, they concluded that there is little or no dose rate effect at low doses from 2500 Gy/h down to dose rates as low as 5 Gy/h. Since our interest is in their EPDM4 (Okonite EPR) which we know is an inverse temperature material, we estimate from their Figure 3 that for room temperature aging, $e/e(0)$ reaches 0.5 after $\sim 5 \times 10^5$ Gy for dose rates ranging from 2530 Gy/h to 14 Gy/h, and a solid horizontal line representing these results is shown on Figure 15b. Also shown on this figure are Sandia results²¹ for Okonite EPR at 40°C + 430 Gy/h and 80°C + 445 Gy/h. It is clear from a combination of the Sandia results and the Virginia results that Okonite EPR is an inverse temperature material. Using thermal-only results generated by Sandia,²¹ the extrapolated thermal-only line at 22°C to $e/e(0) = 0.5$ is also shown on the Figure plus a rough guess (the dashed curve) of what the typical transition from radiation-

domination to thermal domination might look like. Since 150 years at 0.1 Gy/h noted on the Figure is well below the $e/e(0) = 0.5$ estimates, this result would strongly imply that this material would easily survive 60 years at 22°C + 0.1 Gy/h. Again periodic removal of small pieces of cable after 15, 30 and 45 years could be used to reinforce such predictions. Keeping in mind that aging near room temperature is more severe than aging at 50°C for inverse temperature materials, the results just discussed for Brandrex XLPO and Okonite EPR imply that ambient aging at for instance 50°C + 0.1 Gy/h should also create no issues for such materials. Again periodic monitoring could be used to confirm such predictions.

Based on the above discussion, a viable approach for working with inverse temperature materials first requires that combined environment experiments must be done at low temperatures to see if inverse temperature effects are present for a given material. If confirmed, experiments should be conducted versus dose rate at the low temperatures representing ambient aging conditions of interest. By combining these combined environment results with extrapolated thermal aging results at the temperatures of interest and estimating the approximate connections between these two sets of results, one can make reasonable estimates of degradation rates under selected ambient aging conditions. Field-aged samples extracted after selected times (e.g., 15, 30, 45 years) should be considered as a way to gain confidence in the predictions.

The above discussion indicates that many inverse temperature XLPEs and EPRs may be expected to have reasonable mechanical properties after 60 years of worst case ambient aging conditions. It is also clear that attempting to simulate their condition after 60 years using accelerated aging techniques is not currently achievable. This implies that attempts to create simulated 60 year-old nuclear power plant safety-related cables containing these materials to see if they can survive a simulated Loss of Coolant Accident (LOCA) is not currently possible. However, there are reasons for optimism that these materials should be viable under a LOCA. First of all is the fact that the above analyses indicates that these materials should have reasonable mechanical properties after 60 years of ambient aging. In addition, in an earlier publication on inverse-temperature materials, it was observed that oven annealing of materials aged in the inverse-temperature region will often lead to substantial recovery in elongation values.¹⁴ It was concluded that oven annealing leads to a cross-linking repair mechanism at elevated temperatures. For example, a sample of Brandrex XLPO that reached 23% elongation after 328 kGy dose at 22°C plus 200 Gy/h recovered to 225% elongation when placed in a 140°C oven for 24 hours. More detailed studies¹⁴ versus annealing temperatures and times indicate that substantial elongation recovery quickly occurs for this material even at much lower oven temperatures (e.g., recovery to ~150% after ~4 hours at 110°C). Since LOCA environments are postulated to involve an initial day or so of much higher temperatures of around 160°C, an inverse-temperature material that has not degraded sufficiently to cause failure from its ambient aging environment might quickly increase its elongation from cross-linking processes during the first few hours of a LOCA. This implies that the high temperatures involved in any expected LOCA could improve the already reasonable mechanical properties, helping counter the concurrent deleterious effects of the radiation imposed during LOCA. More detailed studies of

such recovery processes for inverse-temperature materials and whether recovery occurs during a LOCA simulation might be helpful in resolving issues of concern for such materials.

CONCLUSIONS

The primary goal of this paper is to emphasize some of the shortcomings that we consider important in several recent IEC and IAEA documents. There is growing evidence that the degradation chemistry in combined radiation-thermal (R - T) environments usually changes with position in (R - T) space given the very different energies involved with radiation initiation (MeV) versus thermal initiation (fractions of an eV). Experimental results for six commonly used cable materials confirm such changing degradation chemistry across (R - T) space. This means there is convincing evidence against the simple time-dependent aging model which is described in the IAEA as well as in IEC documents. As a better approach, we demonstrated that the new MAC version of the DED model can successfully handle such changes in chemistry and can therefore be used to successfully simulate ambient aging conditions for many important cable materials. Because of the expected and observed changes in chemistry across (R - T) space, the “Simplified Method” introduced by JNES¹⁰ and described in the 2012 IAEA document⁹ is also not a viable approach for simulating ambient conditions. The power law model also has important issues. Thus of the four postulated approaches for carrying out accelerated aging in combined $R + T$ environments (power law, time-dependent, DED/MAC and Simplified Method), we conclude that only the DED/MAC approach is broadly useful. Inverse temperature materials, however, present additional difficulties even for the DED/MAC approach, which means a newly suggested method based on extensive low-temperature dose rate experiments reported by Reynolds²⁰ may allow more confident predictions for such materials.

ACKNOWLEDGEMENTS

Sandia National Laboratories is a multi-mission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA-0003525. Any subjective views or opinions that might be expressed above do not necessarily represent the views of the U.S. Department of Energy or the United States Government.

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