SAND2020-11029C

ISSUES WITH APPROACHES FOR SIMULATING AGING OF NUCLEAR POWER PLANT CABLE MATERIALS IN IEC AND IAEA DOCUMENTS

198TH TECHNICAL MEETING OF THE RUBBER DIVISION
ACS INTERNATIONAL ELASTOMER CONFERENCE
KNOXVILLE, TN, OCT. 21, 2020

Kenneth T. Gillen (retired)
Mathew Celina, Organic Materials Org. 1853
Sandia National Laboratories

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & 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-NA0003525.

BACKGROUND

From 1996 to 2017, the International Electrotechnical Commission (IEC) and the International Atomic Energy Agency (IAEA) have published 5 technical documents describing recommended methods for carrying out accelerated radiation plus temperature aging of cable materials in important nuclear power plant environments

FOUR METHODS HIGHLIGHTED

1. Time-dependent model
2. Dose to Equivalent Damage (DED) approach
3. Power law method
4. Simplified method

GOALS OF THIS PRESENTATION

1. Show that the DED approach and a recent modification (the MAC approach) are the only justifiable methods of acceleration for most materials
2. Indicate a possible approach for inverse-temperature materials (E.g., EP, XLPE/XLPO) where the DED/MAC approaches are not viable

APPROACHES FOR ACCELERATED CONDITIONS TO SIMULATE AMBIENT AGING

TIME-DEPENDENT - assumes same degradation chemistry throughout (*R*-*T*) space (same degradation shape when degradation parameter plotted versus log time or log dose)

Insufficient because degradation curve shapes change due to changing chemistry

SIMPLIFIED METHOD - always uses 100°C + 100 Gy/h for accelerating conditions

Fails because *R* is accelerated more than *T* (causing miss-matched chemistry)

POWER LAW APPROACH - assumes at constant *T* that DED ~ R^n

Fails because assumption is typically incorrect

DED AND MAC APPROACH - uses equal acceleration of *R* and *T* that handles changing chemistry

DED described in the literature multiple times and MAC recently described

We believe 3 of the 4 existing approaches have shortcomings

EXISTING GUIDELINES AND RECENT MATCHED ACCELERATED CONDITIONS (MAC) AGING REFERENCES

These documents discuss the Power Law, the Time-Dependent and the DED approaches:

IEC 1244-2 (1st Ed., 1996) and IEC 61244-2 (2nd Ed, 2014): "Determination of Long-term Radiation Ageing in Polymers- Part 2: Procedures for Predicting Ageing at Low Dose Rates", 1996 and 2014.

IAEA-TECDOC-1188: "Assessing and Management of Ageing of Major Nuclear Power Plant Components Important to Safety: In-containment Instrumentation and Control Cables Volume II", Annex E, 2000.

IAEA Nuclear Energy Series No. NP-T-3.6: "Assessing and Managing Cable Ageing in Nuclear Power Plants", 2012. *Also introduces Simplified Method.*

Additionally:

IAEA TECDOC-1825: "Benchmark Analysis for Condition Monitoring Test Techniques of Aged Low Voltage Cables in Nuclear Power Plants", 2017. *Focuses on Condition Monitoring.*

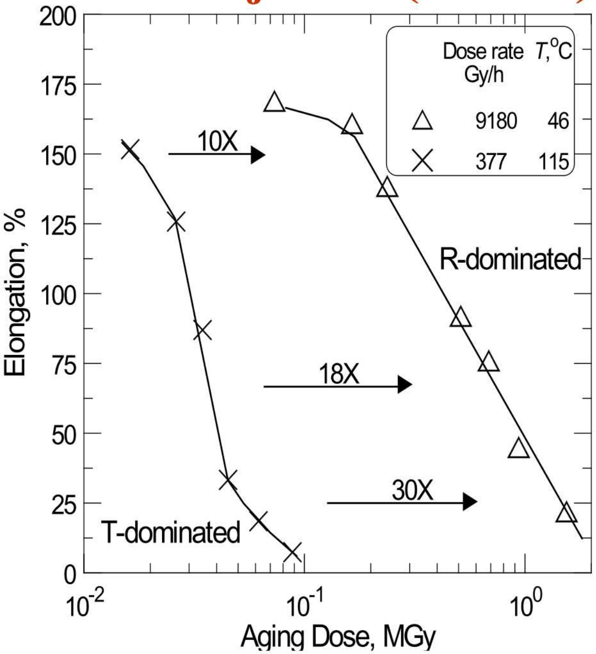
JNES-SS-0903: T. Yamamoto and T. Minakawa, "The Final Report of the Project of "Assessment of Cable Aging for Nuclear Power Plants", July (2009).

K. T. Gillen and M. Celina, "Predicting Polymer Degradation and Mechanical Property Changes for Combined Rad-Thermal Environments", Rubber Chem. Technol., 91, 27 (2018).

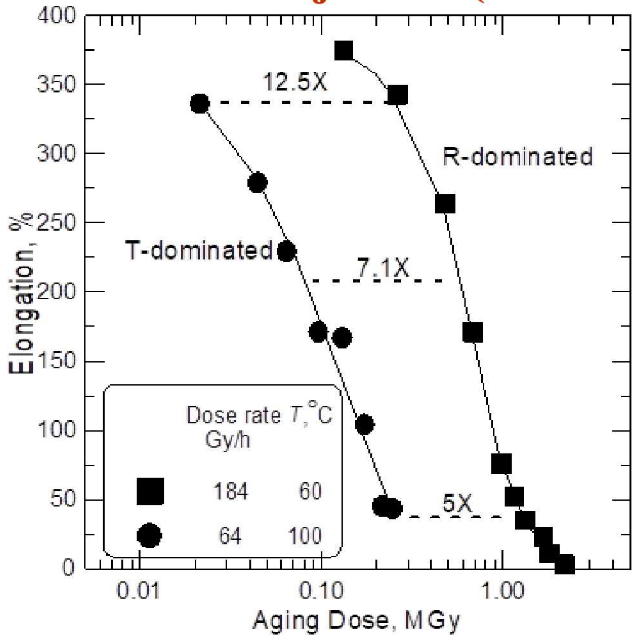
TIME-DEPENDENT MODEL

- **Changing degradation chemistry within *R-T* space challenges *t*-dependent model.**
- **This model requires the same degradation chemistry across *R-T* space (same shape for degradation parameter versus log dose or log time).**
- **<u>Changes are expected:</u> Energy involved in *R*-initiation (1.17 MeV & 1.33 MeV for ^{60}Co) is >> *T*- initiation (0.025 eV at 20°C, 0.036 eV at 150°C). Such changes are experimentally found.**

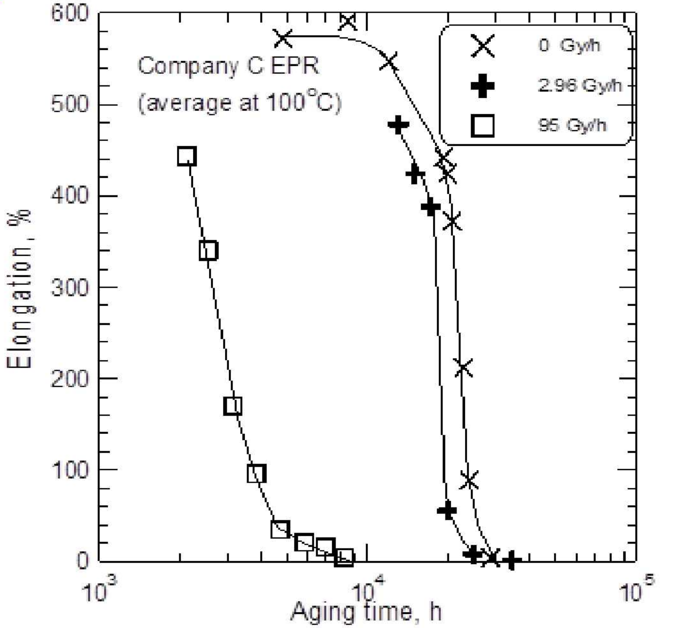
CP cable jacket (Sandia)



CSPE cable jacket (Sandia)



EPR cable insulation*



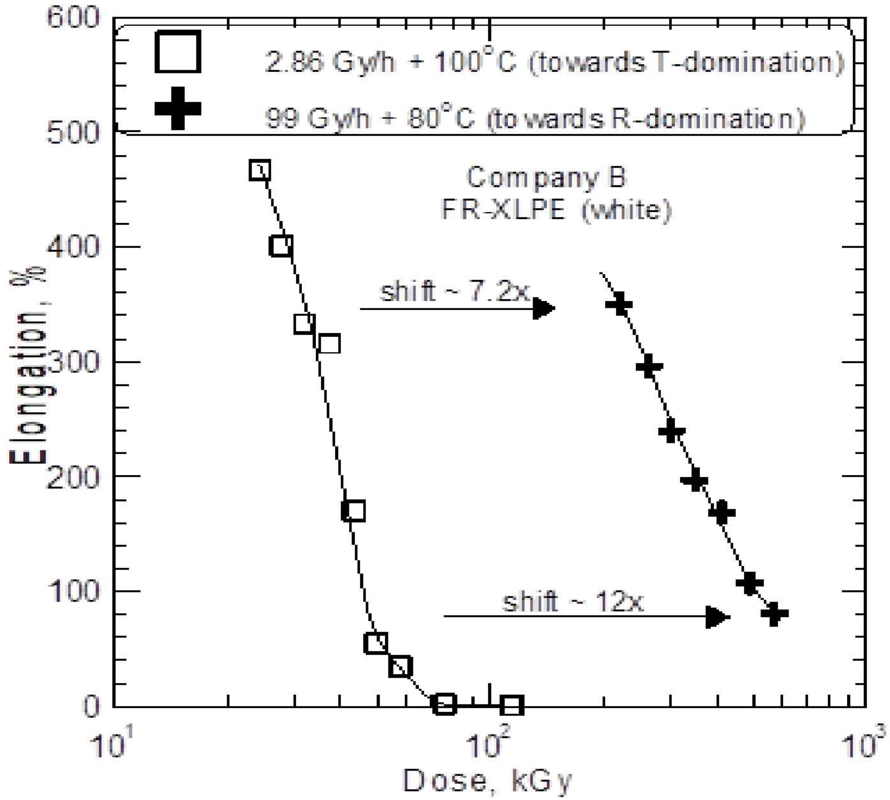
* *T. Yamamoto and T. Minakawa, JNES-SS-0903, July (2009).*

Shape changes in degradation curves imply mechanistic changes

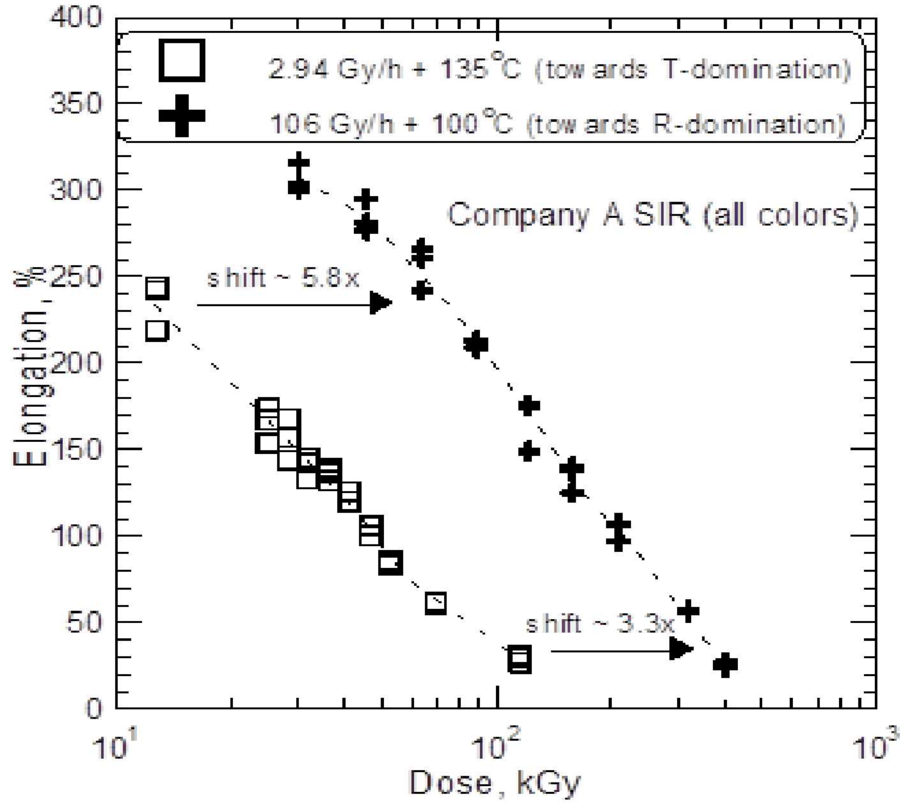
TIME-DEPENDENT MODEL CONTINUED

- **Shape changes are expected and found for most materials examined across *R-T* space.**
- **This implies changes in degradation chemistry, making the t-dependent model inappropriate.**

FR-XLPE cable insulation*



Silicone rubber cable insulation *



* *T. Yamamoto and T. Minakawa, JNES-SS-0903, July (2009).*

Time-dependent model is not valid for changing chemistry

DED AND MAC APPROACH

- **<u>Dose to Equivalent Damage (DED) approach</u> and the recent <u>Matched Accelerated Condition (MAC)</u> update handle changes in chemistry across *R-T* space.**
- **The DED approach is based on the time-temperature-dose rate superposition method which assumes that if the thermal 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 increased by the same factor x (through an increase in dose rate by x), the combined environment degradation rate (plus any synergism) will increase by the same factor x. DED usually looks at a given degradation amount (e.g., elongation reaching 50%) whereas MAC examines the entire degradation curve.**

DED APPROACH REFERENCES

1. K. T. Gillen and R. L. Clough, "A Kinetic Model for Predicting Oxidative Degradation Rates in Combined Radiation-Thermal Environments", J. Polym. Sci., Polym. Chem. Ed., 23, 2683 (1985).
2. 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", Polym. Degrad. Stab. **24**, 137 (1989).
3. K. T. Gillen and R. L. Clough, "Accelerated Aging Methods for Predicting Long-Term Mechanical Performance of Polymers", in **Irradiation Effects on Polymers,** D. W. Clegg and A. A. Collyer, Eds., Elsevier Applied Science, London, 1991., Ch. 4.

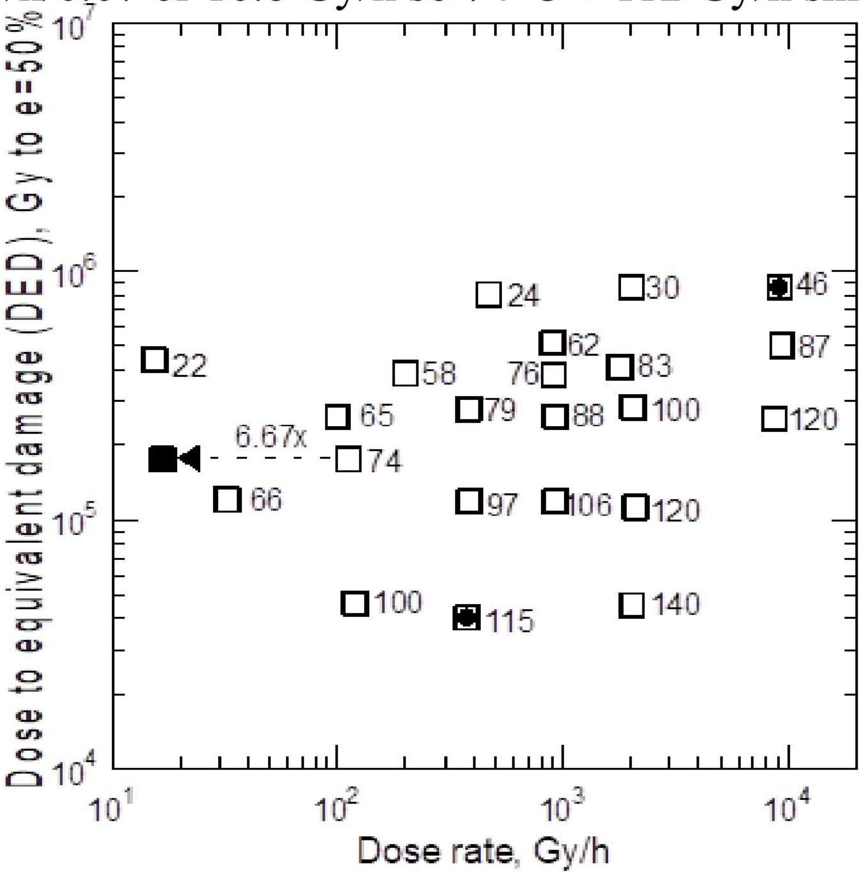
MAC APPROACH REFERENCE

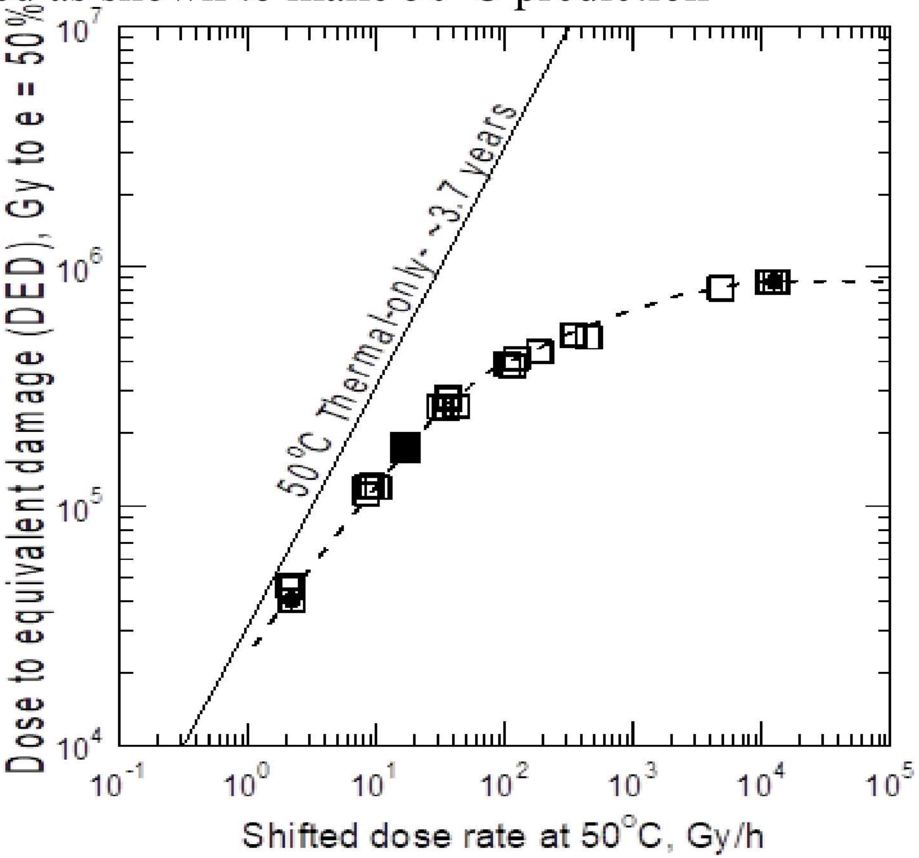
K. T. Gillen and M. Celina, "Predicting Polymer Degradation and Mechanical Property Changes for Combined Radiation-Thermal Environments", Rubber Chem. and Technol., **91**, 27 (2018).

The DED and MAC approach allows for changing chemistry

DED EXAMPLE

22 *R* + *T* conditions for a CR cable jacketing material to reach 50% elongation. Arrhenius factor from 74°C to 50°C is 6.67. DED model assumes that data taken after time, *t* at 74°C + 112 Gy/h is equal to data taken after time, 6.67*t* at 50°C + 112 Gy/h/6.67 or 16.8 Gy/h so 74°C + 112 Gy/h shifted as shown to make 50°C prediction





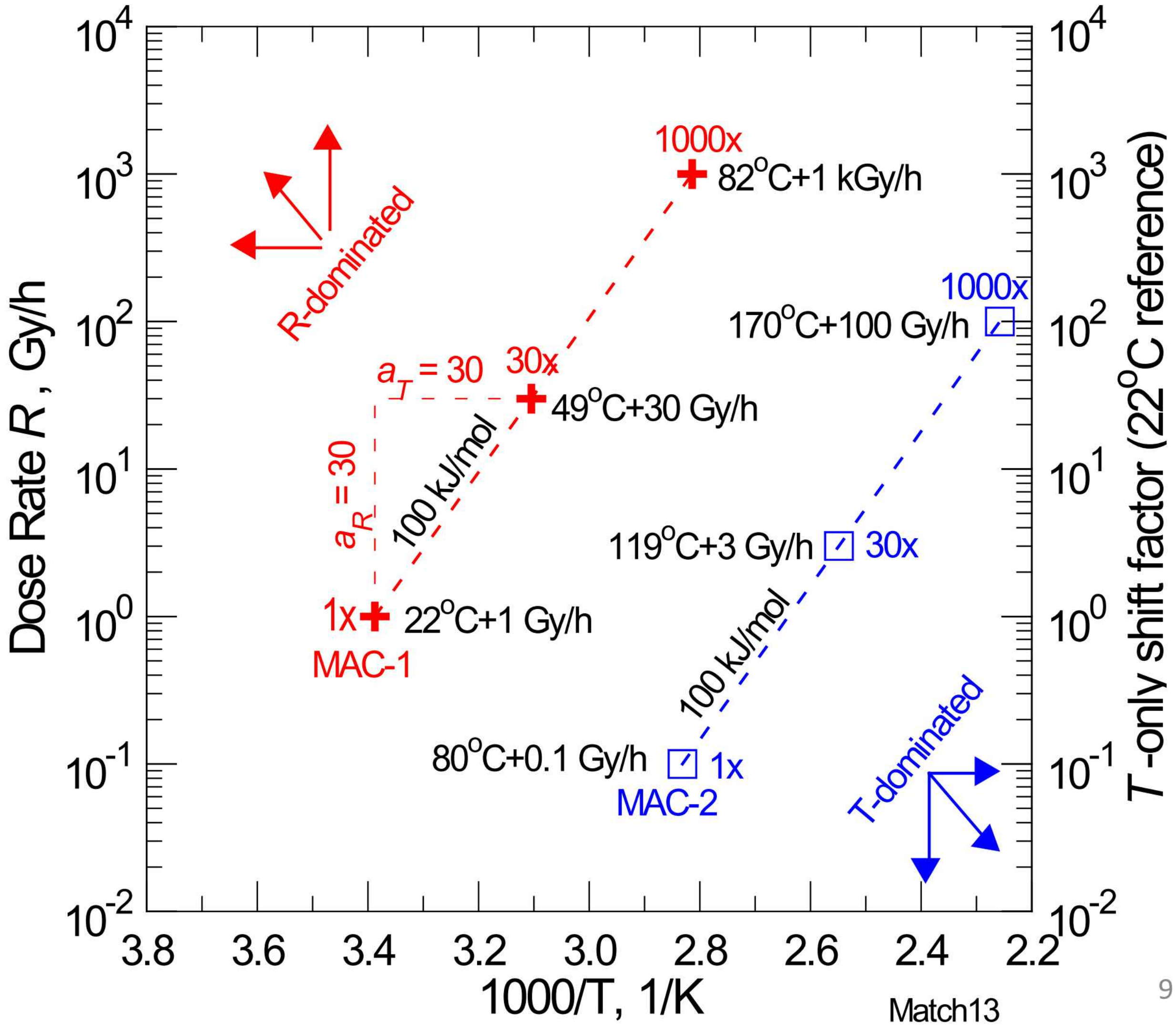
Superposition from 22 data points shifted according to assumptions yields one degradation curve and results versus dose rate reflect chemistry changes but only to *e* = 50%. Could analyze at differing elongation values to explore how dose-dependent curves change with dose rate.

New DED model approach based on analysis under Matched Accelerated Conditions (MAC)

Simulate 22°C + 1 Gy/h and E_a is 100 kJ/mol for *T*- only aging. Using this E_a we can calculate *R* + *T* conditions for various accelerations. Points along this line are referred to as Matched Accelerated Conditions (MAC).

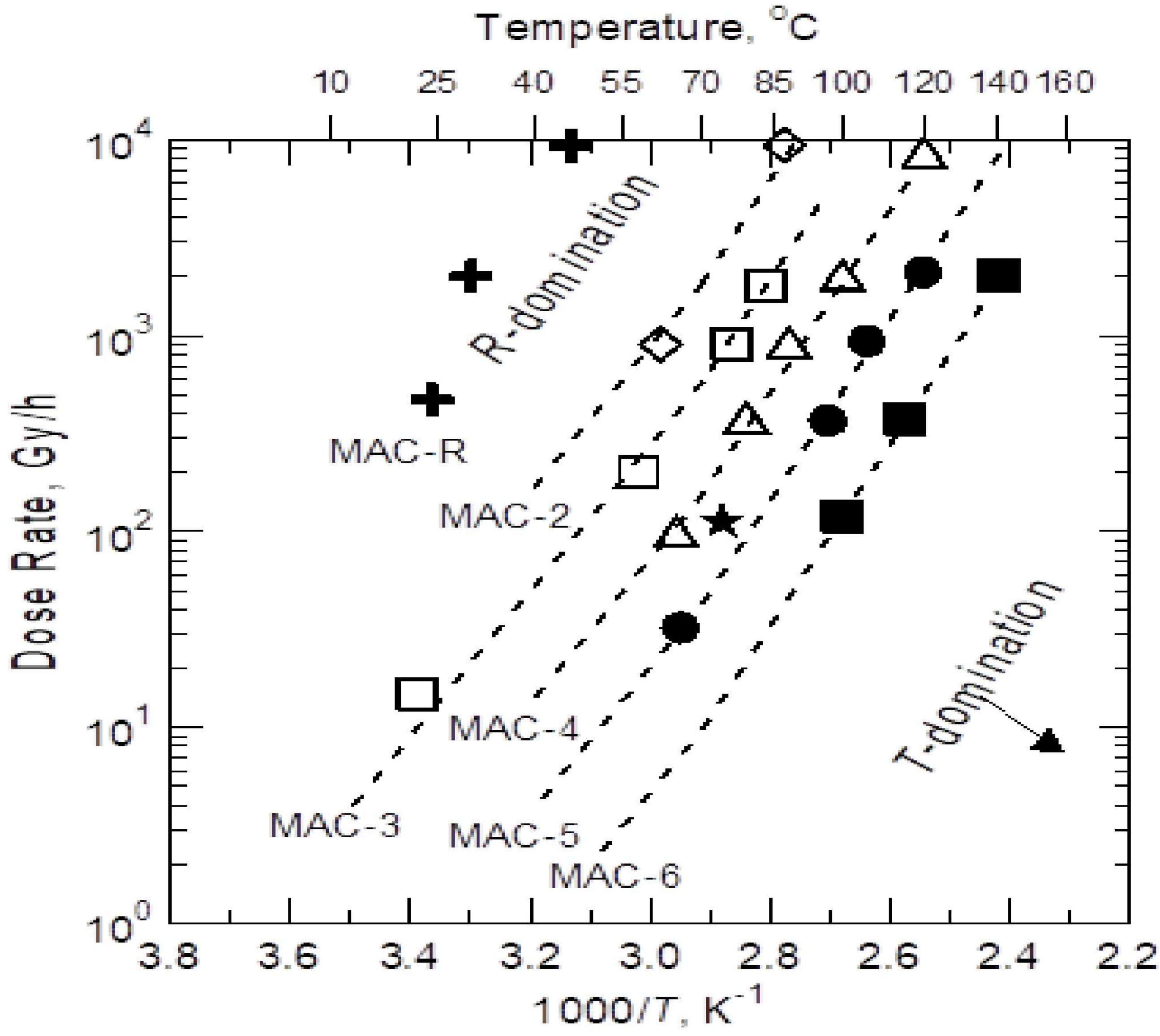
If data taken under differing conditions on a MAC line superpose versus dose, this confirms assumption that increases by a factor x in thermal initiation (from its activation energy) and a similar increase by a factor x in dose rate will increase the overall degradation rate by the same factor x.

Changing chemistry can be handled



MAC APPLIED TO CR CABLE JACKET

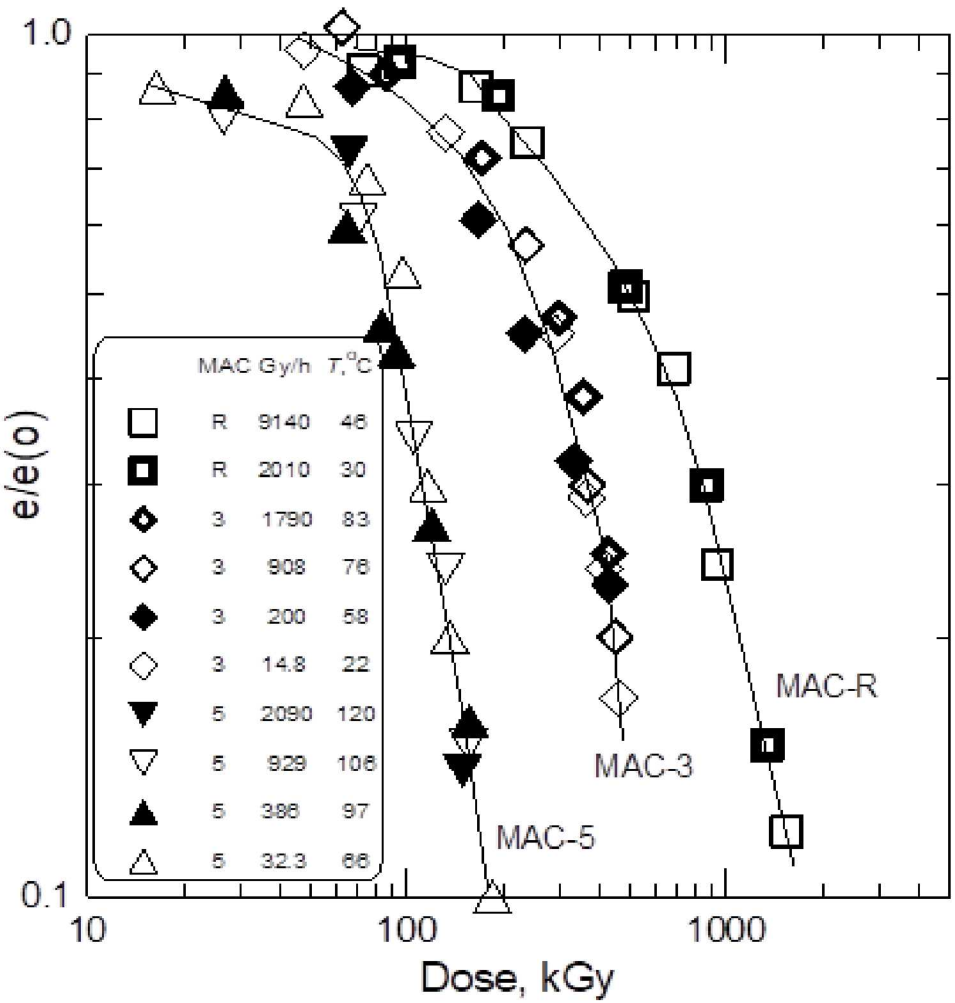
22 data locations planned to be along 6 MAC lines from *R*-domination to *T*- domination (probing chemistry changes)

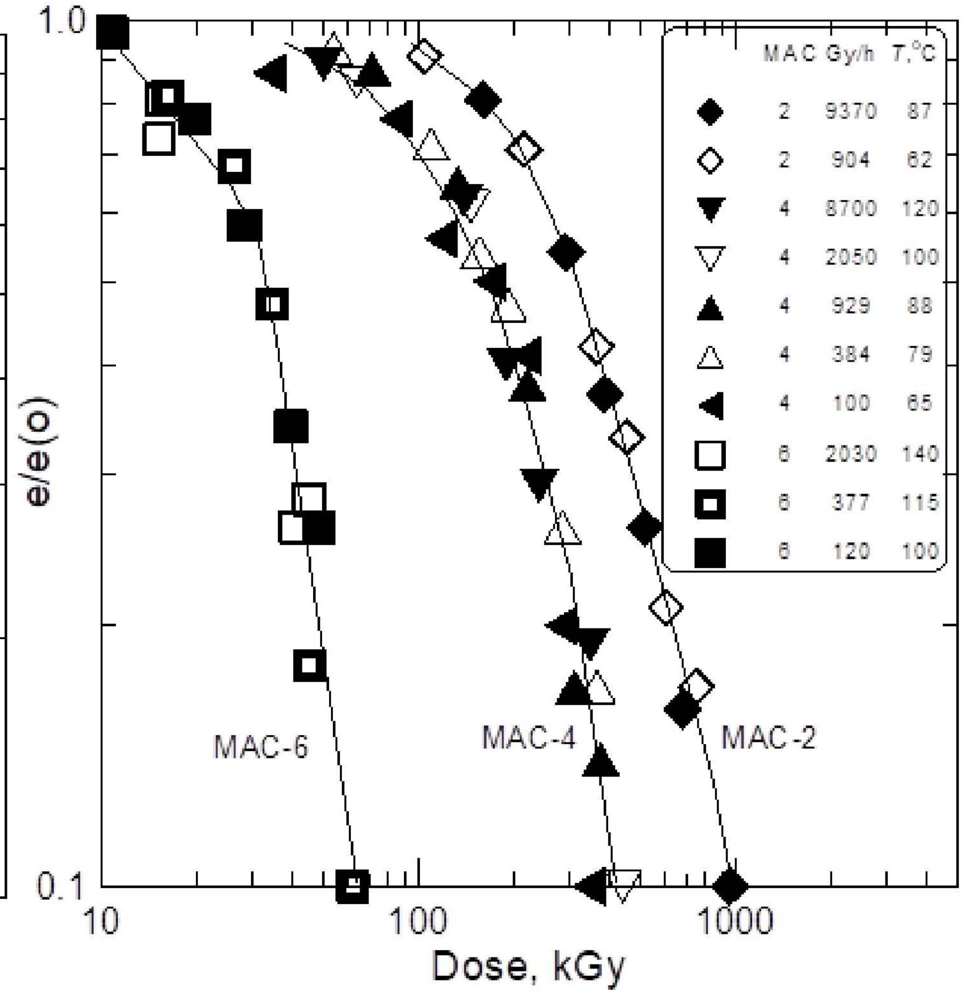


Along a MAC line, DED model predicts all data should superpose when plotted versus dose

NICE SUPERPOSITION FOR ALL 6 MAC LINES

This confirms model assumptions. Note the large accelerations involved (~120x for MAC-3 and ~87x for MAC-4).



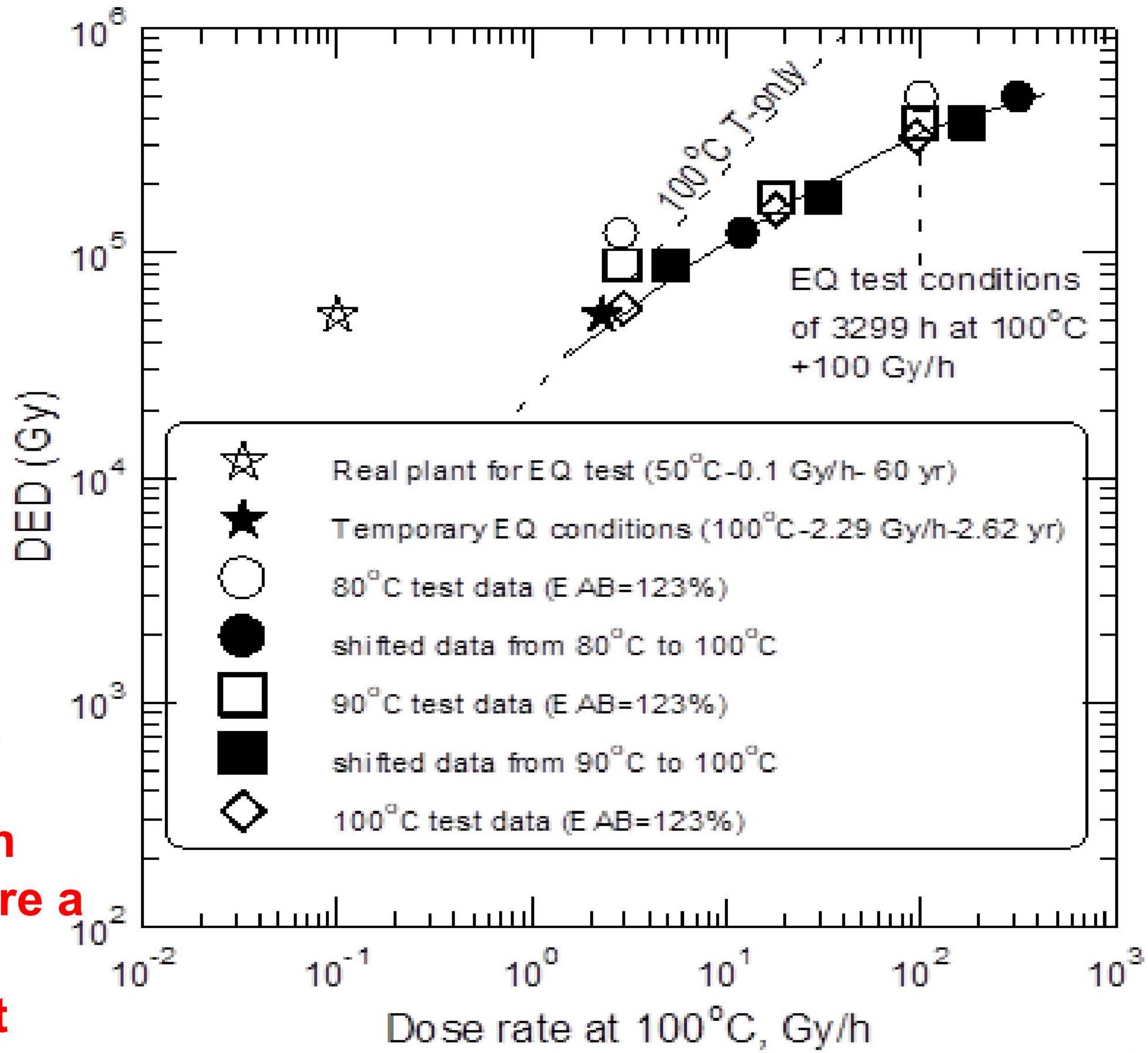


Results reflect degradation chemistry changes across *R-T* space
Each MAC line represents slightly different chemistry

SIMPLIFIED METHOD APPROACH JNES-SS-0903, July (2009)

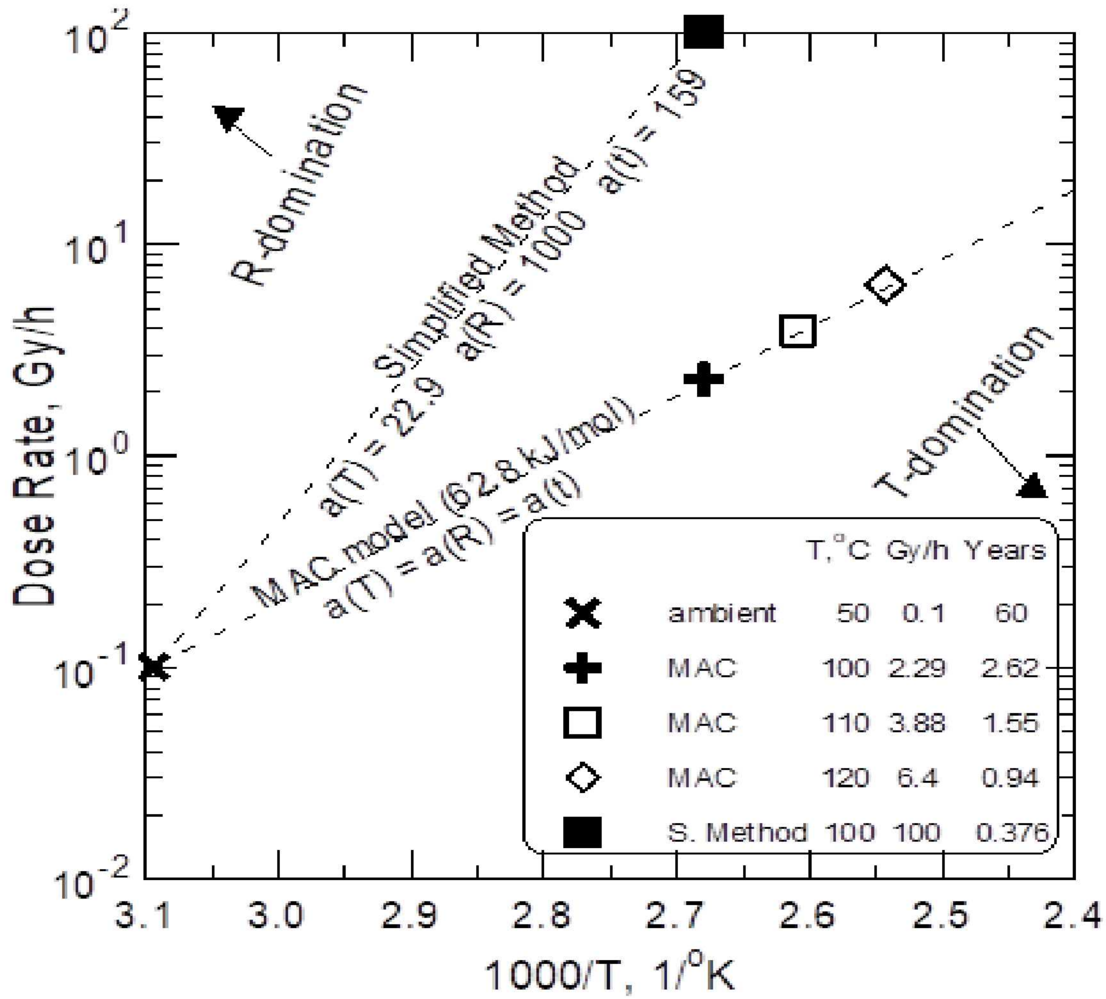
Involves a <u>*complex*</u> 6-step procedure (highlighted in the 2012 IAEA document). The accelerated conditions are always 100°C + 100 Gy/h.

For instance for the EPM insulation from Company C with ambient aging conditions of 60 years at 50°C + 0.1 Gy/h and an assumed activation energy of 62.8 kJ/mol, the procedure results in accelerated aging conditions of 3299 h at 100°C + 100 Gy/h.



Unfortunately the chosen accelerated conditions are a total miss-match of the chemistry under ambient aging conditions

SIMPLIFIED METHOD APPROACH- CONTINUED



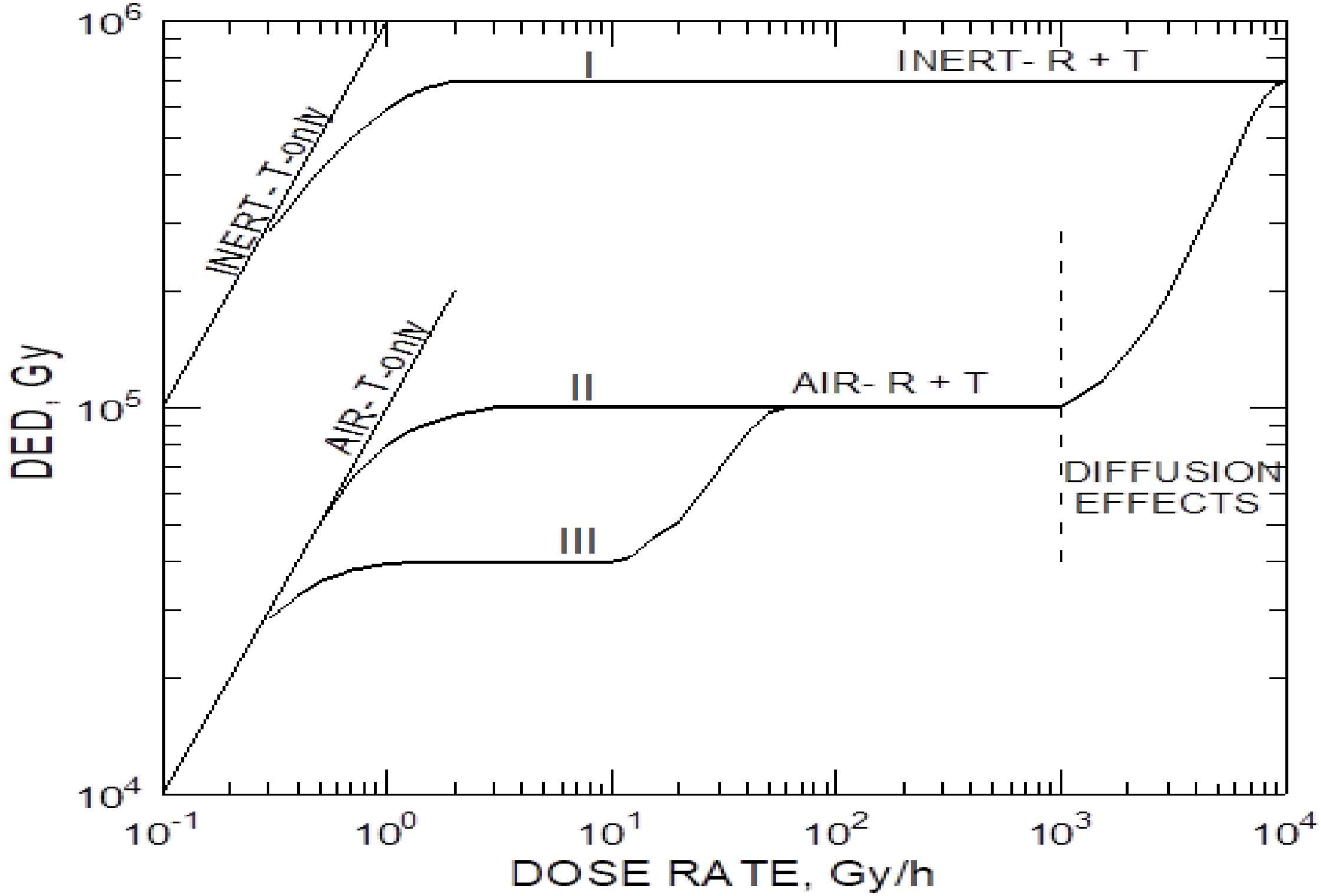
Ambient and accelerated conditions plotted on a MAC-type plot.

Assuming E_a is accurate, the proper accelerated aging conditions would be along the MAC line (equal acceleration of *T, R* and time, *t*. The JNES method accelerates *T* by 22.9, *R* by 1000 and *t* by 159, thereby moving the chemistry much closer to radiation domination.

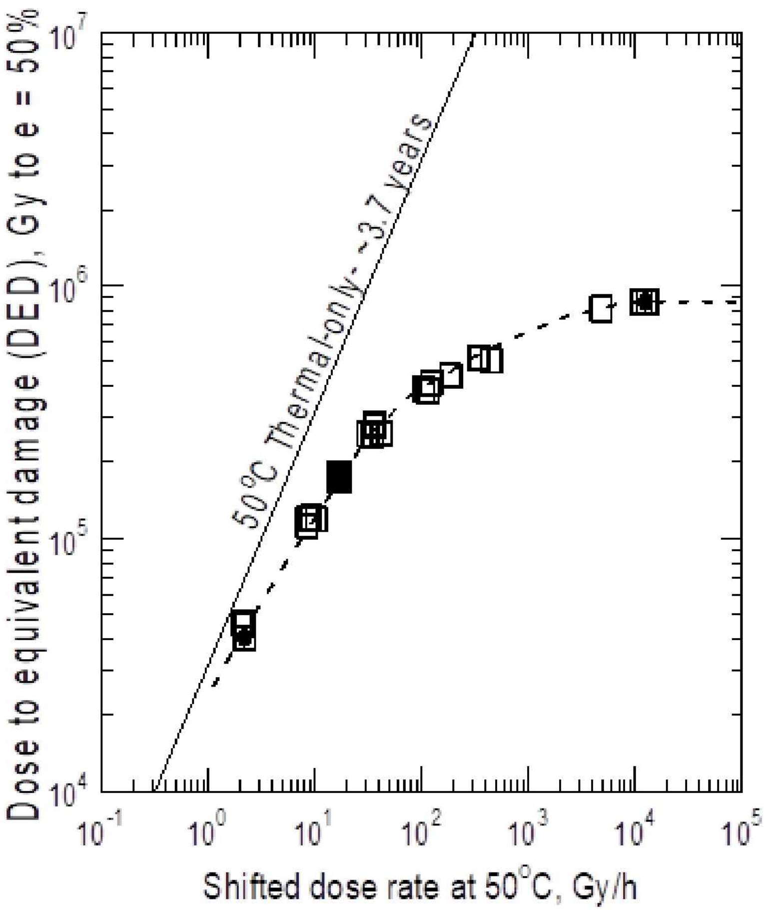
Accelerated conditions do not reflect the chemistry occurring under ambient aging conditions

POWER LAW APPROACH

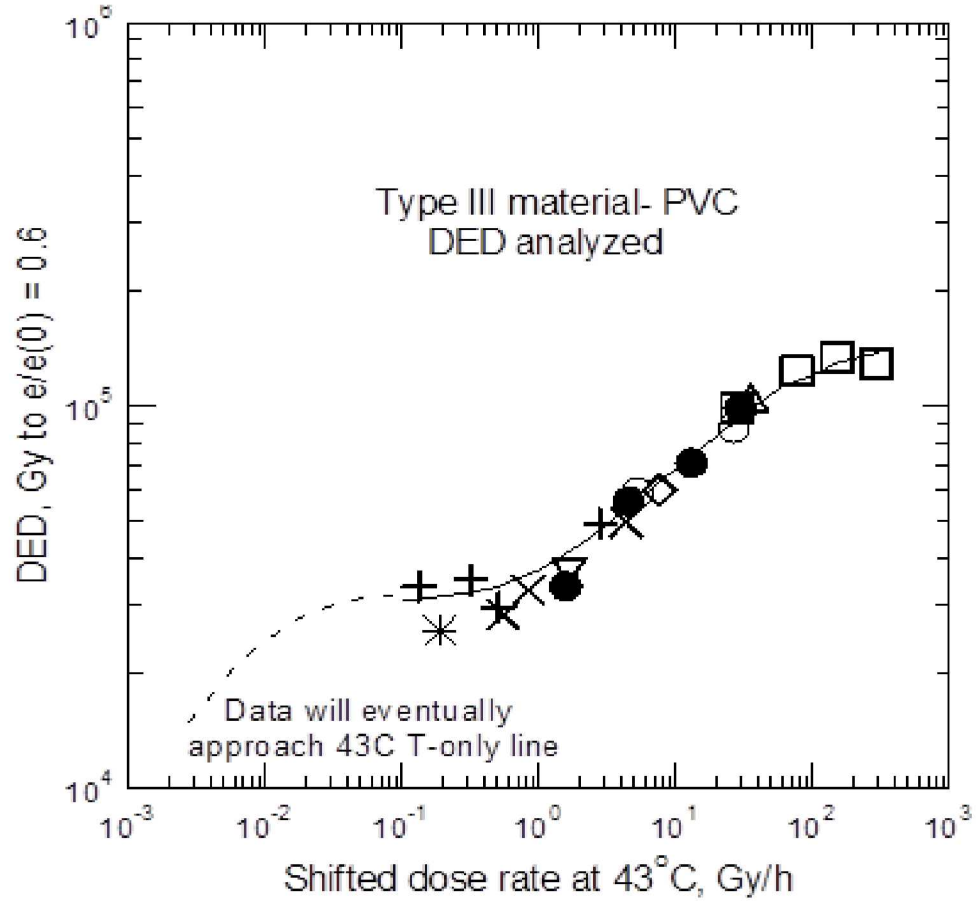
- **Predicts a linear relationship between log of DED and log of *R* at constant aging *T* conditions.**
- **Countless experimental results over the past 40 years show that almost all materials under air aging follow Type II or Type III behaviors shown in this figure**.



TYPE II- CR CABLE JACKET



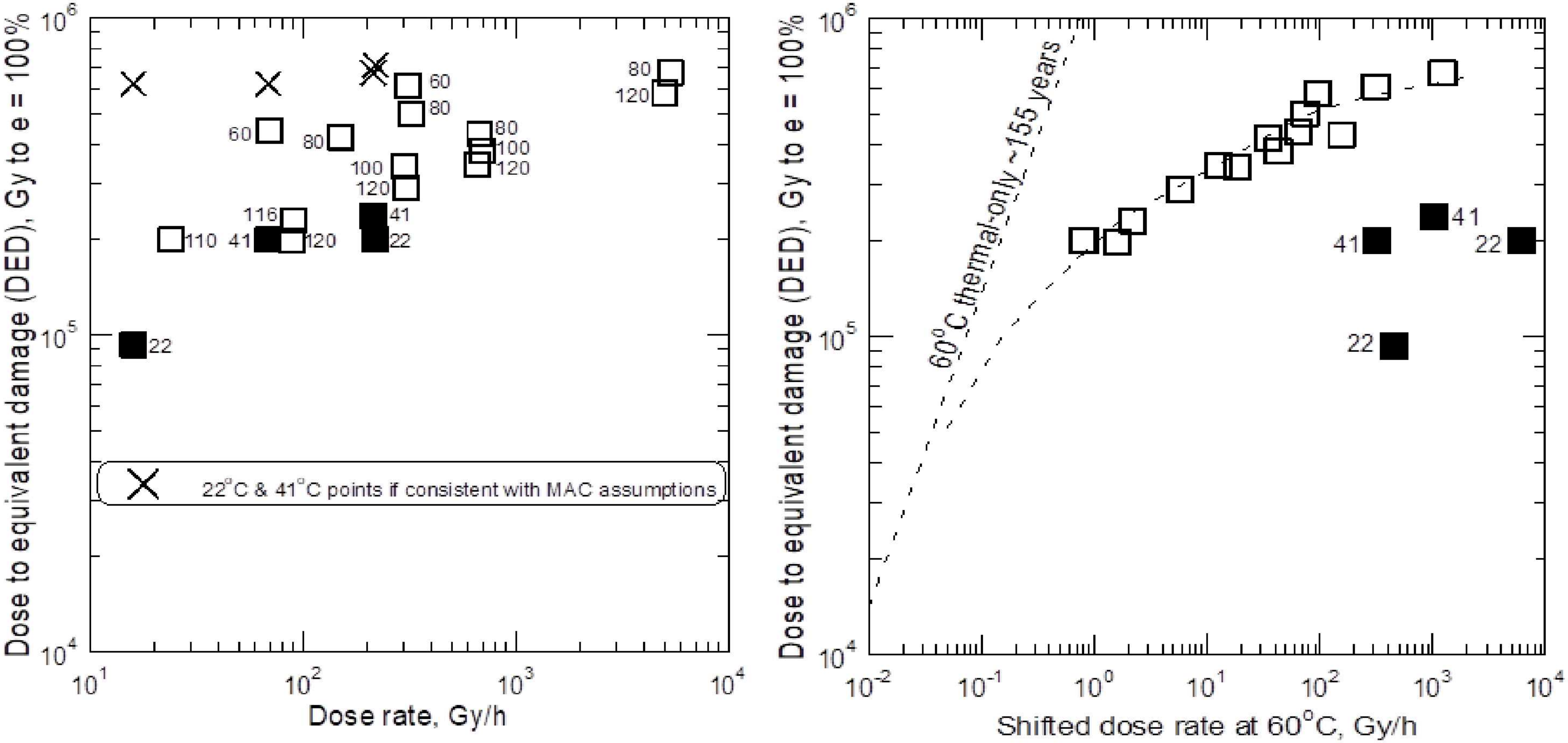
TYPE III- PVC JACKET



Therefore power law approach is hard to justify
Further details are provided in Conference paper

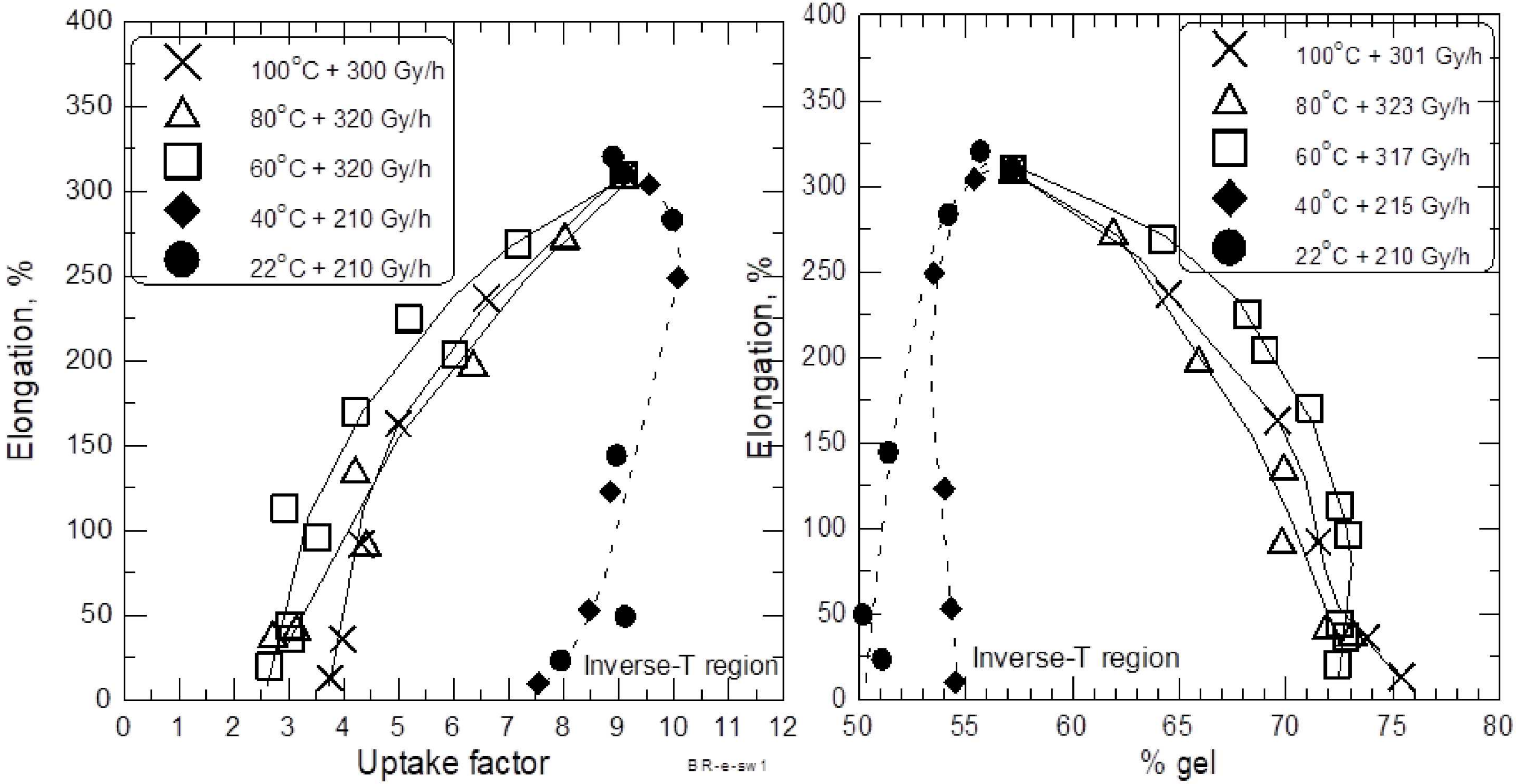
INVERSE TEMPERATURE COMPLICATIONS

- **Many EP and XLPE/XLPO materials have degradation rates increase at constant *R* when *T* drops below ~60°C**
- **Example- Brandrex XLPO cable insulation material where DED analysis shows problems**



Inverse temperature means 'degradation chemistry' is convoluted by physical material behavior

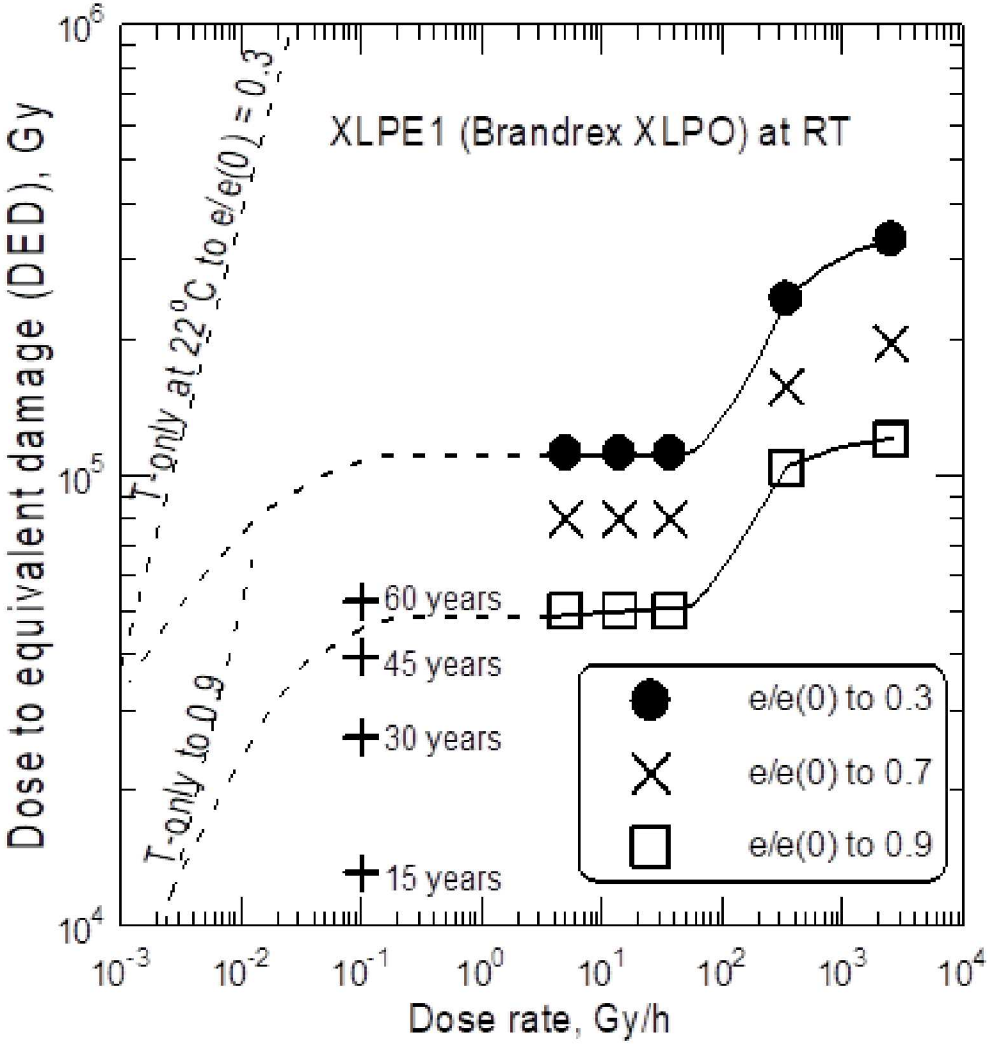
DRASTIC CHEMISTRY CHANGES OCCUR IN INVERSE-T REGION



Scission becomes much more important in inverse-*T* region
Therefore aging must be done under ambient *T* conditions

WORK UNDER LOW T CONDITIONS DONE FOR MANY INVERSE TEMPERATURE MATERIALS BY REYNOLDS

A. B. Reynolds, R. M. Bell, N. M. N. Bryson, T. E. Doyle, M. B. Hall, L. R. Mason, L. Quintric and P. L. Terwilliger, Radiat. Phys. Chem., **45**, 103 (1995).



- **Increase above ~100 Gy/h due to DLO**
- **Leveling out below 100 Gy/h will eventually transition to *T*-domination limit (Type II behavior)**
- **At 0.1 Gy/h, 60 years of aging is unlikely to reach low elongation values.**

Periodic removal of small pieces of cable for testing every 15 years or so could reinforce such predictions

CONCLUSIONS

- **Initiation energy differences (MeV for *R*, fractions of an eV for *T*) imply degradation chemistry changes vs. position in *R-T* space**
- **Experimental results confirm such changes in degradation chemistry**
- **These results argue against time-dependent method**
- **The DED approach and its MAC update successfully handle changes in chemistry**
- **The Simplified Method has significant shortcomings since it accelerates *R* much greater than *T***
- **The power law approach is inconsistent with expected dose rate dependence at constant *T***
- **A suggested approach might be useful for inverse-*T* materials**
- **Thus of the 4 methods described in IEC/IAEA documents, only the DED and recently improved DED-MAC approach is viable for accurate predictions at service conditions**