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TECHNICAL REPORT

Determination of long-term radiation ageing in polymers –
Part 4: Effects of different temperatures and dose rates under radiation conditions

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INTERNATIONAL
ELECTROTECHNICAL
COMMISSION

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DETERMINATION OF LONG-TERM RADIATION AGEING IN POLYMERS –

Part 4: Effects of different temperatures and dose rates under radiation conditions

FOREWORD

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IEC TR 61244-4, which is a Technical Report, has been prepared by IEC technical committee 112: Evaluation and qualification of electrical insulating materials and systems.

The text of this Technical Report is based on the following documents:

Draft TR	Report on voting
112/442/DTR	112/446/RVDTR

Full information on the voting for the approval of this Technical Report can be found in the report on voting indicated in the above table.

This document has been drafted in accordance with the ISO/IEC Directives, Part 2.

A list of all parts in the IEC 61244 series, published under the general title *Determination of long-term radiation ageing in polymers*, can be found on the IEC website.

The committee has decided that the contents of this document will remain unchanged until the stability date indicated on the IEC website under "http://webstore.iec.ch" in the data related to the specific document. At this date, the document will be

- reconfirmed,
- withdrawn,
- replaced by a revised edition, or
- amended.

A bilingual version of this publication may be issued at a later date.

INTRODUCTION

IEC 60216 (all parts) and IEC 60544 (all parts) give reference and guidance for managing accelerated thermal and radiological ageing steps for type testing procedures applicable to electrical insulating materials. The actual application of electrical equipment usually requires the consideration of effects which are a consequence of simultaneous occurrence of temperature and radiation at varying intensities.

The CIGRE WG D1.42 study presents degradation data in particular with respect to cable and wire insulation materials gathered from tests where thermal and radiation loads were applied simultaneously. Even if there is a broad range of materials available from the industry, only insulation materials commonly used were selected for this study. These materials are crosslinked polyethylene (XLPE), ethylene-propylene-rubber (EPR), silicon-rubber (SIR) and polyvinylchloride (PVC). Using these test data, power plant operators were in the position to meet requirements defined by regulatory bodies in the frame of 'long term operation application', showing that most insulation materials which have been in operation for 30 to 40 years were in good condition. Furthermore, material samples were collected from real positions and test results were compared with reference samples, unaged as well as artificially aged.

The main objective of the industry is to yield reliable values of the residual lifetime of the insulation materials and linked pieces of equipment made up of these materials. However more research is necessary as the in-service degradation of insulating materials appears to be deviating from estimation based on accelerated ageing tests. For a better determination of the degradation processes of insulation materials it is important to gain a wider knowledge on material degradation and linked synergistic effects at low intensities of thermal and radiological loads. Thus, this document aims to summarize the results, and in some areas update the literature references, from CIGRE WG D1.42, to provide a state-of-the-art document on qualification procedures capable to represent multifactor ageing (hereby thermal and radiological ageing).

DETERMINATION OF LONG-TERM RADIATION AGEING IN POLYMERS –

Part 4: Effects of different temperatures and dose rates under radiation conditions

1 Scope

This part of IEC 61244 provides general guidance for the evaluation/verification of electrical insulation materials (EIM) and electrical insulation systems (EIS) intended to be used in types of equipment exposed to ionizing radiation. Beside sensors, actuators/motors as well as plugs and terminals, cables are a well-known typical application of those EIM and EIS. Their type spectrum covers low voltage power cables, control cables and instrumentation cables. Because of their comparable simple design, cables are the ideal type of equipment to study EIM and EIS degradation processes. But the results of these studies can be easily transferred to the enumerated types of equipment.

Nonetheless, this document provides a state-of-the art report on qualification/verification procedures used to simulate simultaneous effects of temperature and radiation at varying intensities rather than give detailed test programmes valid for specific test methods.

NOTE 1 Use of this document with specific products can require specification of additional product related procedures.

NOTE 2 Some of the procedures described in this document are emerging technologies. Therefore, specified prerequisites, former experiences as well as boundary conditions can be additionally taken into account.

2 Normative references

The following documents are referred to in the text in such a way that some or all of their content constitutes requirements of this document. For dated references, only the edition cited applies. For undated references, the latest edition of the referenced document (including any amendments) applies.

IEC 60544 (all parts), *Electrical insulating materials – Determination of the effects of ionizing radiation*

IEC TS 61244 (all parts), *Determination of long-term radiation ageing in polymers*

3 Terms, definitions and abbreviated terms

3.1 Terms and definitions

For the purposes of this document, the terms and definitions given in IEC TS 61244 (all parts) and IEC 60544 (all parts) apply.

ISO and IEC maintain terminological databases for use in standardization at the following addresses:

- IEC Electropedia: available at http://www.electropedia.org/
- ISO Online browsing platform: available at http://www.iso.org/obp

3.2 Abbreviated terms

Abbreviated term	Definition
ACA	assessment of cable ageing for nuclear power plants
AO	anti-oxidant
CEA	Commissariat à l'Energie Atomique
CERN	European Organization for Nuclear Research
CRIEPI	Central Research Institute of Electric Power Industry
CSPE	chrolo-sulphonated polyethylene
DED	dose to equivalent damage
DLO	diffusion-limited oxidation
DOR	Department of Operation Reactor
DSC	differential scanning calorimetry
EAB	elongation at break
EdF	Electricité de France
EIM	electrical insulation materials
EIS	electrical insulation systems
EPDM	ethylene propylene diene rubber
EPR	ethylene propylene rubber
EPRI	Electric Power Research Institute
EQ	environmental qualification
FR	flame retardant
HDPE	high density polyethylene
HELB	high energy line break
IAEA	International Atomic Energy Agency
IEEE	Institute of Electrical and Electronics Engineers
IEEJ	Institute of Electrical Engineers of Japan
IH	inhibitor (antioxidant)
IR	infrared
IRSN	Institut de Radioprotection et de Sûreté Nucléaire
JAEA	Japan Atomic Energy Agency
JAERI	Japan Atomic Energy Research Institute
JAMPSS	Japan Ageing Management Program on System Safety
JNES	Japan Nuclear Energy Safety Organization
LDPE	low density polyethylene
LET	linear energy transfer
LOCA	loss of coolant accident
MSLB	main steam line break
NISA	Nuclear and Industrial Safety Agency

Abbreviated term	Definition
NPP	nuclear power plant
NRC	Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
PE	polyethylene
PVC	polyvinyl chloride
QST	National Institutes for Quantum and Radiological Science and Technology
SIR (SiR)	silicone rubber
SNL	Sandia National Laboratories
TED	time to equivalent damage
TMI	Three Mile Island
XLPE	cross-linked polyethylene
XLPO	cross-linked polyolefin

4 Radiation induced degradation mechanisms at standard ambient conditions

4.1 General conventions

The effects of radiation on polymer materials have been studied in both material reformation and degradation since the 1950s [1-3][1]. Specific attention has been paid to polymer materials used for electrical insulation and jackets of cables in the 1960s, where commercial nuclear power plants were designed and built extensively [3]. The first results describing the behaviour of insulation materials were reported from ORNL [4-6] and Harwell Atomic Energy Research Establishment [7].

Secondary electrons can be generated in the bulk material due to the photoelectric effect, the Compton effect, etc., when polymer materials are exposed to ionizing radiation. These electrons can induce molecular ionization and electronic excitation [8]. Such physical phenomena were believed to be the main cause of material degradation at that time, because radiation penetrated well into the bulk material. It was also assumed that the contribution of radiation to degradation was far greater than from heat for the same reason [9]. Accelerated degradation through the presence of oxygen was also known, but this factor was underestimated in fundamental research. Chain scission was considered to occur only in the presence of oxygen, and cross-linking was assumed in non-oxygen atmospheres [3,10].

These early studies focused on the physical effects of radiation such as collision cross-sections and linear energy transfers (LET) [3]. All studies of academic interest were thought to be completed in 1967 [2]. However, the importance of oxidation had still not been identified and most experiments were conducted in vacuum or under an inert atmosphere. As a result, dose rate effects [9] were not considered and accelerated ageing experiments were performed at high dose rates based on the "equal dose equal damage" concept. Permissible total radiation dose [11] was adopted to define the radiation resistance of polymers. Such databases can be found in various reports from ORNL [4-6], CERN [12-15], JAEA (current QST) [16], and EPRI [17] and in many other literatures [18-25].

1 Numbers in square brackets refer to the bibliography.

The ‘equal dose equal damage’ concept was also reflected in the IEEE type test standard [26] for qualification of safety-related equipment in nuclear power plants, and accelerated ageing with dose rates up to 10 kGy/h was accepted. The over dose approach, which uses a greater amount of radiation than the in-service simulated total dose, was also used at this time to improve maintainability. This approach was also used to compensate for uncertainties in the knowledge at the time [27]. SNL strongly emphasized that these tests did not take into account dose-rate effects [28].

4.2 Effect of presence of oxygen

Radiation is one source of radical generation and chemical bond breaking, and heat may generate radical, while heat assists oxidation reactions. Material degradation occurs via synergistic effects of radical generation and oxidation based on an auto-oxidation scheme. Mechanical properties such as tensile strength do not exhibit significant degradation [29] under irradiation in an inert gas environment. EAB, resistivity, and dielectric loss (tanδ) do not change when irradiation occurs in N_2, whereas significant adverse effects for these properties are observed when irradiation occurs in air [30]. Furthermore, infrared (IR) absorption due to carbonyl groups is small immediately after a material is irradiated, but the IR peak would be increased by subsequent thermal ageing in a sequential accelerated ageing test. This generation of carbonyl groups correlates with decrease in EAB [29]. Seguchi et al. have reported that 80 % to 90 % of oxygen that contributes to oxygen reactions would be accumulated in the bulk, and not less than 80 % of that forms carboxylic acid [10].

4.3 Effect of dose-rate effect-1: Physical aspects

It is widely known that the DED value decreases as the dose rate decreases, and this phenomenon is called a dose-rate effect, irrespective of the causes. Put in other words, whilst strong radiation induces severe material degradation, if total dose is used as the base, a lower radiation environment can lead to more severe degradation [31] at a given total dose.

Such dose-rate effects were identified as early as 1956 at the basic research level [6, 22, 32-34]. Wilski et al. have pointed out that inhomogeneous degradation in bulk was the cause [35-37]. Gillen et al. experimentally clarified the relation between the inhomogeneous degradation and decrease in EAB [38,39]. This inhomogeneous degradation is caused by DLO, which occurs if the oxidation reaction rate is faster than the oxygen supply from the outside air and its diffusion into the bulk of the material. Inhomogeneous degradation is also observed in a high-temperature environment even if no radiation is present [40]. Gillen et al. also quantitatively demonstrated that the rate of oxygen supply from air is decreased with oxidation, which results in enhancing DLO in the subsequent degradation process [40]. Further advancements in the characterization of DLO and its effect on lifetime predictions under accelerated thermal ageing was provided in a seminal paper by Gillen et al. [41].

Dose-rate effects in a fluoro-rubber has been investigated [42] and found that degradation processes (embrittlement or flexibility) are dependent on radiation dose-rates. Therefore, accelerated ageing tests that give rise to inhomogeneous degradation were suspected to simulate the ageing process in service. It has been commonly recognized that accelerated ageing tests should be carried out under conditions that do not generate DLO. Limits of test conditions that give homogeneous degradation have been already summarized elsewhere; one of the examples listed in IEEE Std 775 [43] is shown in Table 1. Conditions to prevent DLO can also be investigated by analytical calculation as detailed by Gillen et al. [41].

Table 1 – Dose rate conditions which do not cause inhomogeneous degradation

Material	**Dose rate** Gy/h			
	Sheet thickness			
	0,5 mm	1,0 mm	1,5 mm	2,0 mm
HDPE	50	13	5,8	3,2
LDPE	440	110	49	27
EPR	4 800	1 200	530	300
EPDM	2 100	520	230	130
Hypalon	1 200	300	130	75
Neoprene	520	130	58	32
SIR	5 100	1 300	580	320
PVC	440	110	49	27

NOTE Irradiation temperature is 25 °C [43].

4.4 Effect of dose-rate-2: Chemical aspects

It is clear that lifetime or so-called TED would show a constant value when the radiation dose-rate is low enough that the thermal oxidation reaction is dominant for material degradation. Since constant TED values induce a ‘thermal line’ in DED curves, these exhibit so-called chemical dose-rate effects [39,44], to be distinguished from the DLO-derived physical dose-rate effects.

Initially, this chemical dose-rate effect was not recognized. Gas analysis, frequently used at the beginning of investigations, showed that oxygen absorption increased with a decrease in dose rate [37]. The oxidation reaction was not accelerated when the oxygen concentration was increased [10], although there is sparse supporting data for this observation at present. It has also been reported that the chemical dose-rate effects were no longer observed if polymeric materials are stabilized by inhibitors [45]. IEEJ technical report [11] recognized the importance of dose-rate effects only after such phenomena were confirmed by long-term tests performed at dose-rates as low as several tens of mGy/h in the US and France [46,47] or by EQ tests on XLPO cables [48].

4.5 Research on degradation mechanism

Chemical reaction is considered as the mechanism of degradation in insulating polymeric materials since the above-mentioned two types of dose rate effects have been established and the importance of oxygen recognized. Further, irradiated materials are observed to degrade without exhibiting an induction period if they are subsequently heated [28]. Degradation factors appear to accumulate in the bulk during irradiation. Peroxide species (ROOH) are reported as such factors; for example, thermal decomposition of ROOH would promote an auto-oxidation chain reaction, resulting in thermal degradation of PE [49-52]. Clough et al. have found that removing ROOH by phosphine gas suppresses degradation [53].

It is also reported that radiation ageing followed by thermal ageing for sequential accelerated ageing generates rapid degradation, even when one to six months have passed between radiation and thermal ageing. The accumulated degradation factor is considered to be stable ROOH because radicals directly generated by radiation are generally difficult to sustain for such a long period. Activation energy (E_a) values obtained for EAB measurements [54,55] coincide with that for ROOH decomposition [29,53]. This also suggests that ROOH is the rate-controlling factor in material degradation, which also supports the use of an auto-oxidation scheme that encompasses ROOH.

Fuse et al. have recently proposed a new model with taking into account the influence of antioxidants (see 4.15), of various temperature effects and divides the effects of thermal and radiation on radical formation. This work highlighted the need for further work in these areas to refine their auto-oxidation model [56].

4.6 Experiences and acceleration factors

Attention was drawn to the ability of safety-related equipment to continue to function during LOCA conditions by surveys disclosed to the public by the mid-1980s [11]. Cables are simultaneously exposed to high-temperature steam and strong radiation in the LOCA environment, so attention was focused on synergistic effects in this complex environment [3]. The order effect is synergistic and was an issue related to the sequential type test, as indicated in the phase of basic studies. Consideration of these effects lead to concerns that the accelerated ageing test conditions selected would have resistance against LOCA conditions. Acceleration factors, which are roughly the ratio of type test time to the assumed actual in-service period, has been recognized to be too high since at least 1990. This concern is addressed in 4.7 below.

4.7 Low dose-rate tests and LOCA survivability

The US NRC, SNL and CEA have studied the effects of accelerated test conditions on LOCA durability, by exposing various cables to mild irradiation conditions from 5 Gy/h to 100 Gy/h and temperatures from 40 °C to 70 °C for up to five years [46]. One of the results showed that accelerated ageing tests give rise to material degradation, and that the subsequent LOCA test further degrades functional characteristics. Most cables investigated maintained insulation functions even after the LOCA test, but one of the XLPE cables degraded excessively and failed.

Concerns about the resistance to LOCA conditions against material degradation due to low dose rate irradiation were raised by several SNL studies [57-59]. The US NRC pointed out the possibility that insulations of multi-conductor cables swelled from jackets during the LOCA period, and the swelling point could be the failure point. The study further concluded that cables with a jacket bonded to the insulation provide low durability against LOCA [60]. In the US, Wyle Laboratories aged insulating materials with an equivalent of 60 years of in-service exposure and found that increase in leakage current and dielectric breakdown strength occurred in some cables after the LOCA test. Based on this finding, the US NRC reported that extending cable life might increase degradation of the material [60].

Germany and France conducted long-term irradiation tests over ten years [61,62]. EPRI has also conducted a long-term irradiation test and installed sample cables in nuclear plants that were also evaluated as part of their studies [61], [63-65]. No property changes due to in service ageing have been found for these samples, but the evaluation is still underway. CERN's long-term test results are summarized in the literature [66,67].

4.8 Effect of LOCA test environment

Equipment to perform simultaneous heat and radiation irradiation called SEAMATE-II was built in former JAERI Takasaki in 1979 [47], [68-71] to verify the equivalence of the simultaneous and sequential methods. The US also conducted similar studies and reported no differences between the two kinds of testing methods in the case of LOCA conditions [58]. This result was attributed to the environments in which the test objects were irradiated; cable samples were irradiated under oxygen-free conditions in the simultaneous method, whereas irradiation in the sequential method was performed in air. This difference in atmosphere derives from the process that oxygen-decomposed species are expelled from autoclave equipment together with compressed steam and oxygen [72]. Thus, it was reported that the sequential method provided a more severe environment. The effect of oxygen concentration during LOCA tests was also investigated in the above-mentioned NRC/SNL/CEA joint research [46]. The results indicated that oxygen concentration did not affect the performance decrease in XLPE, ethylene tetrafluoroethylene, SIR, EPR, and FR-XLPE, although performance was affected in the case of chloroprene rubber. The research has suggested the necessity to evaluate the temporal change in the oxygen concentration in containment during a LOCA period. It was also reported that material degradation tends to be alleviated under high humidity [3]. Chemical spray used as a coolant in the pressurized water reactor LOCA test was reported to have small effect on material degradation compared to γ-ray irradiation or steam temperature [3,47,73,74].

4.9 Order effects, synergistic effects, and antagonistic effects

As has already been discussed above, radiation induces radical formation, which is the ignition process of chemical reactions, whereas heat may generate radicals. It follows that degradation is enhanced by the reverse (backward) sequential method in which radiation is performed in advance, rather than by the normal sequential method, at least qualitatively. The optical absorption of carbonyl groups that is frequently used as a degradation index [28] remains small under irradiation at room temperature, but increases by thermal degradation thereafter [29]. However, material degradation is also greatly affected by specimen shape, manufacturing process, material, composition, humidity, oxygen, temperature, etc. As a result, order effects in sequential tests and thermal and radiation synergistic effects were not recognized for some time. Synergistic effects were confirmed with CSPE but not with EPR in the literature [46]. As these two kinds of radiation effects have gradually been observed in many other materials, both have been considered to be generated by the same mechanism [38].

Order effects also affect the type test standardized by IEEE, since this standard does not specify the test sequence [53]. Concerns about the reproducibility of in-service environment by sequential methods would also arise in the case that degradation behaviour exhibits strong synergistic effects [29]. The issue becomes complex in a high-temperature ageing environment, because heating would also take part in radical generation. Clarifying the reaction rate and its activation energy (E_a) against relevant elementary chemical reactions is necessary to quantitatively evaluate the contribution of heat and radiation but would be difficult.

The technical reports published by IEEJ [11,47] have recognized from studies at SNL [75] that degradation in many polymeric materials becomes more severe in the reverse (backward) sequential method than in the forward method. The report [11] has also mentioned that irradiation should be performed first in order to follow the NRC DOR guidelines [76]. It is also worth noting that stress factors under in-service environment are not yet completely understood, and that difficulties still need to be overcome for the present ageing test procedures to provide lifetime estimation [11]. An accelerated ageing test is considered as a qualitative screening technique [11].

In a series of experiments evaluating the lifetime evaluation of NPP cables a reverse (backward) sequential (radiation then thermal) ageing combination was found to be more severe than the more common sequential order (thermal then radiation). The simultaneous ageing was found to be in between these two approaches in terms of severity. This work highlighted the importance of oxidation and the influence of antioxidants in these results [77].

4.10 Sequential test conditions equivalent to simultaneous methods

The biggest issue, yet to be resolved, in synergistic and order effects is to identify test conditions for the reverse (backward) sequential ageing test equivalent to those in simultaneous methods. Literature [78,79], has recommended carrying out heat ageing after irradiating materials at 70 °C to simulate degradation by the simultaneous method. It is also effective to irradiate with a dose rate of 650 Gy/h and then perform heat ageing at 120 °C, or to irradiate materials with 1,2 kGy/h and heat at 140 °C. The maximum acceptable dose rate to induce homogeneous oxidation within EPR and XLPE materials was found to depend on irradiation temperature. The temperature influences the rate of diffusion of oxygen into the material as noted in 4.3. Hence, a dose rate of 50 Gy/h was found to be acceptable for 1 mm thick XLPE at room temperature. This maximum dose rate increased to 800 Gy/h if the irradiation took place at 100 °C. Much higher values were obtained for EPR material which exhibits a higher oxygen diffusion rate. However, the higher irradiation temperature can also induce additional chemical reactions which can impact the ageing mechanisms. Based on these findings the irradiation temperature should not exceed 100 °C [80].

4.11 Studies after TMI and later

There have been many long-term studies investigating low dose-rate radiation since the early 1990s. This research was prompted by the standardization of several time-temperature-dose rate superposition methodologies which include the one named 'superposition of DED data' by IEC and IAEA. These two organizations have also standardized evaluation and monitoring methods for material degradation, due to the concern over nuclear accidents (e.g. Chernobyl). As long-term irradiation tests require years for evaluation, the amount of research has gradually decreased. In the case of Japan, the Nuclear Technology Assessment Committee of the Ministry of Economy, Trade and Industry designated evaluation technology for cable degradation as one of the urgent implementation items in ageing management in 1999. The project called 'Assessment of Cable Ageing for Nuclear Power Plants (referred to as ACA)' was performed by JNES [81].

The objective of the ACA research was to validate the Japanese national standard for type testing, in response to international trends in low dose-rate accelerated ageing procedures. Therefore, low dose-rate irradiation was performed on safety-related cables used in Japanese NPPs. As a result, it was found necessary to bend the Arrhenius line in order to correlate in-service cable conditions to test results obtained between 100 °C and 130 °C. Studies up to then had focused on radiation effects and temperature characteristics which assumed the normal Arrhenius law. This bending called 'non-Arrhenius behaviour' was also approved by SNL [82]. However, SNL also suggested that the ACA research reconsider the following two points in their evaluation. First, the use of tentative activation energy value (E_a) with respect to polymeric materials in which the in-service data did not indicate significant degradation. Second, no inverse temperature effect was taken into account with respect to crystalline polymers like XLPE and EPR.

4.12 Arrhenius law and limitations

Non-Arrhenius behaviour has also been recognized on the basic research level, similar to the case for other irradiation effects described above. IEEJ technical reports [11,47] stated, with respect to Arrhenius behaviour, that XLPO has a linear characteristic against thermal life whereas EPR exhibits 'poor' linearity. JAEA also presented related studies in 1984 [9,83]. The Arrhenius law is based on the assumption that the activated molecule concentration and the molecular collision probability increase at a constant rate with increase in temperature resulting in promotion of chemical reactions. Consequently, the ease of reaction changes, and nonlinearity occurs, if ageing tests exceed any temperatures such as glass transition temperature and/or melting point, no matter if they are of the base material or of additives related to degradation.

The E_a values are also important together with other governing factors with respect to non-Arrhenius behaviour. IRSN reported that in-service cable degradation takes place more quickly than predicted by a conventional linear Arrhenius plot. This finding agrees with that reported in the ACA project [81]. IRSN expressed concern that the uniform use of the E_a value had a limited applicable range [84]. Table 2 summarizes the E_a values reported. Both the E_a values and the temperature that exhibits the nonlinearity vary in materials, manufacturers, and grades.

Table 2 – E_a value according to materials, manufacturers, and grades

Manufacturer[a]	Grade	Low temperature		High temperature		Organization	Reference
		E_a kJ/mol	Temperature °C	E_a kJ/mol	Temperature °C		
XLPE							
Rockbestos	2/C #14 AWG			127	180 to 200	NRC	63
Brandrex	CLPO	72	50 to 144	72	50 to 144	SNL	82
ITT	Surprenant Exane II			110	90 to 130	SNL	82
Unknown	Unknown	54 to 79	60 to 78	110	90 to 170	SNL	35, 85
Unknown	Unknown			128	Unknown	EPRI	86
Company A	CV	(61 to 78)		110	100 to 120	JNES	84
Company B	CV			100	100 to 120	JNES	81
Company C	CV			95	100 to 120	JNES	81
Company A	FR-CV			66	100 to 120	JNES	81
Company B	FR-CV			84	100 to 120	JNES	81
EPR							
American Insulated Wire	3/C #16 AWG			119	141 to 160	NRC	9
Anaconda	Flameguard 14AWG			106	100 to 170	SNL	87
Anaconda	Durasheeth 12AWG			100	99 to 139	SNL	87
Eaton Dekoron	Elastoset 16AWG			106	110 to 160	SNL	82
Okonite	12AWG			89	100 to 140	SNL	82
Unknown	Unknown	75	< 110	115	110<	SNL	88
Unknown	Unknown			116	Unknown	EPRI	86
Unknown	Unknown	58	< 120	126	120 <	JAEA	89
Company C	EP rubber			94.5	100 to 120	JNES	81
Company A	FR-EPR, black			110	100 to 120	JNES	81
Company A	FR-EPR, white			102	100 to 120	JNES	81
Company A	FR-EPR, red			96	100 to 120	JNES	81
Company B	FR-EPR, black			84	100 to 120	JNES	81
Company B	FR-EPR, white			85	100 to 120	JNES	81
Company B	FR-EPR, red			87	100 to 120	JNES	81

Manufacturer[a]	Grade	Low temperature		High temperature		Organization	Reference
		E_a kJ/mol	Temperature °C	E_a kJ/mol	Temperature °C		
Company C	FR-EPR, black			87	100 to 120	JNES	81
Company C	FR-EPR, white			84	100 to 120	JNES	81
Company C	FR-EPR, red			85	100 to 120	JNES	81
SIR							
Company A	KGB			47	135 to 175	JNES	81
Company B	KGB			42	135 to 175	JNES	81
Company C	KK			50	135 to 175	JNES	81
Chlorosulfonated polyethylene (Hypalon)							
Anaconda	Flameguard inner			107	< 100	SNL	87
Anaconda	Flameguard outer			107	< 100	SNL	87
Kerite	FR			107	< 100	SNL	87
Rockbestos	Firewall III	88	40 to 100	107	< 100	SNL	87
Eaton Dekoron	Elastoset			107	< 100	SNL	87
Brandrex	Unknown			107	< 100	SNL	87
BIW	Bostard 7E	88	82 to 100	107	< 100	SNL	87
Samuel Moore	Dekoron	88	40 to 100	102	100 to 125	SNL	87
Unknown	Unknown	80 to 105	22 to 125	80 to 105	22 to 125	SNL	90
Unknown	Unknown	22	> 120	57	< 120	JAEA	91
Mitsubishi Cable	Unknown	55	> 120	97	< 120	Mitsubishi Cable	61
Unknown	Unknown			80 – 96	unknown	NRC	9
Unknown	Unknown		> 100		< 120	EdF	92
Chloroprene (Neoprene)							
Okonite	Unknown	89	24 to 70	71	70 to 131	SNL	82
Burke Rubber	Unknown (w/filler)			86 to 98	65 to 125	SNL	93
Burke Rubber	Unknown (w/o filler))	37	25 to 66	98	66 to 140	SNL	94
Unknown	Unknown	75 to 100	22 to 115	75 to 100	22 to 115	SNL	90
Unknown	Unknown	71	> 70	89	< 70	SNL	88
Unknown	Unknown	82	> 80	96	< 80	SNL	88
Polyvinyl							

Manufacturer[a]	Grade	Low temperature		High temperature		Organization	Reference
		E_a kJ/mol	Temperature °C	E_a kJ/mol	Temperature °C		
chloride							
Unknown	Unknown			84 – 96	43 to 110	SNL	90
Nitrile rubber							
Burke Rubber	Unknown	75	23 to 52	88	52 to 96	SNL	93
Unknown	Unknown	84	RT to 125	84	RT to 125	SNL	88
Unknown	Unknown	88	RT to 125	88	RT to 125	SNL	88
Butyl rubber							
Unknown	Unknown	75	> 80	100	< 80	SNL	88
Polyurethane							
Unknown	Unknown	70	> 70	120	< 70	SNL	88

[a] This information is given for the convenience of users of this document and does not constitute an endorsement by IEC of the companies named.

4.13 Slow degradation behaviour observed in service-aged cables

The Japanese electric industry has performed condition assessment of cables installed in their nuclear power plants [95], in accordance with the documented direction by NISA [96]. In-service degradation is slower than that expected from the master curve superposition even when non-Arrhenius behaviour is taken into account. If present master-curve shifting is used, higher E_a values are required to reproduce severe degradation in the actual environment. Similar issues have already been pointed out for accelerated ageing on epoxy resins used for instrumentation equipment [97-99]. Non-Arrhenius behaviour seems not only to simply modify the data shift procedures, but also requires some modifications to the chemical reaction scheme in material degradation.

The reaction rate of the auto-oxidation scheme has been considered as a factor in non-Arrhenius behaviour. Some literature [100,101] focuses attention on the chain initiation reaction while others [102-104] focus on the termination reaction. With respect to the latter, Gillen et al. applied gas analysis to chloroprene rubber and successfully measured non-Arrhenius behaviour of E_a against gas consumption and evolution [103]. That is, CO_2 is generated efficiently at high temperatures, whereas this factor is no longer important under low-temperature conditions. Similar trends have been observed for CSPE [105].

CRIEPI has looked at the decrease in antioxidant (referred to as IH, also referred to as AO) and its critical concentration [106,107]. The research carried out under JAMPSS also pointed out the importance of considering the IH effect. Another study is now irradiating several kinds of polymeric materials using extremely mild conditions to study ageing behaviour close to in-service environments using nuclear magnetic resonance and other high-sensitivity techniques [108,109]. However, it is also important to note that chemical techniques also require correlation with functional parameters such as mechanical properties.

A recent study of flame-retardant EPR insulation removed from containment after 16 years in service was in agreement with these observations. The lifetime of the in-service material was shown to be three times longer than the one obtained from accelerated ageing. The difference in behaviour was attributed to suppression of the oxidative reaction and crosslinking upon further ageing of the field material. Control samples were found to display a higher level of chain scission upon ageing [110].

4.14 Inverse temperature effects

Generally, the degradation rate would be lowered with a decrease in temperature. However, the reverse also takes place in the case of high crystallinity of EPR and XLPE. Such materials exposed to temperatures generally 40 °C or lower sometimes degrade extremely rapidly. This phenomenon, where degradation occurs more quickly in a low-temperature environment rather than in a high-temperature environment, is called the inverse temperature effect.

Table 3 shows instrumental analytical results on such behaviours. Chain scission is indicated to be occurring at temperatures 40 °C or lower, however, density and crystallinity rise. This conflicting phenomenon is resolved by annealing. Gillen et al. have assumed that ROOH would play a role with an analogy to the cable degradation model, but experimental results obtained using ROOH decomposing gas showed that this assumption is no longer valid. It is tentatively assumed that tie molecules which take part in bonding crystalline and amorphous regions would engage in chain scission, and that annealing causes re-crystallization and molecular exchange to contribute to the recovery phenomenon [61,111]. The authors of these references recognize that this phenomenon is merely a notation in international reports to date, but many related papers have already reported on it [46,78,82,85,111,112].

Table 3 – Changes in various properties due to inverse temperature and annealing recovery effects, investigated by several instrumental analyses [111].

Property	< 60 °C	20 °C to 40 °C	Annealing effect
EAB	At higher temperature, degradation is fast	Degradation slower than that in 120 °C environment	Recovers to same level as 60 °C degradation
Oxygen consumption rate	Exhibits Arrhenius characteristics	Scarcely exhibits temperature dependence	
Gel fraction	Molecular cross-linking takes place	Molecular chain scission takes place	Molecular cross-linking takes place
Density	No change	Increases	Decreases
Crystallinity	No change	Increases	Decreases
DSC	No change	New peaks appear at 45 °C and 80 °C	

4.15 Role of antioxidants

It has long been recognized that IHs can suppress material degradation [113-118]. Generally, IHs can be divided into the following two types: one is an energy absorption type represented by the aromatic system, and the other is a radical absorption type represented by a hindered phenol system. The former absorbs radiation energy and is deactivated by de-excitation; the latter stabilizes materials by reacting with radicals. Consequently, the former tends to remain in the bulk material and is somewhat difficult to dissipate.

The US NRC has suggested that rapid decrease in EAB would be observed when the IH is depleted from the cable insulation [119]. This phenomenon is known as critical concentration characteristic; degradation would rapidly take place after the IH falls below a certain threshold concentration.

The effectiveness of antioxidants was found to be mostly lost below a critical content of approximately 0,04 weight per cent. Accelerated ageing can affect the influence of antioxidants by increasing their rate of evaporation in comparison to normal ageing conditions. Under these conditions heterogeneous oxidation can also take place which influences the overall properties of the material. These findings also relate to changes in activation energy at higher accelerated ageing temperatures [120].

4.16 Other thermal and radiation environment degradation characteristics

1) The effects of electron beams and γ rays on degradation are the same if the amount of energy absorption is the same [121].
2) It has been pointed out that cables have only one surface exposed to the oxygen atmosphere [11]. Material degradation tends to be mild in the case of cable-shaped samples, not only insulation but also jackets [122].
3) Humidity does not affect material degradation [78], nor do water and humidity accelerate degradation. Any decrease in electrical insulating properties tends to be delayed as compared to decrease in mechanical properties [47]: an effect attributed to vaporization of low molecular-weight substances by heating.
4) SNL advocates that flame retardants do not affect material degradation [123]. On the other hand, IEEJ technical report [11] mentioned that there are various types of degradation behaviour in FR-type EPR and XLPE related to the addition of flame retardants.
5) Pigments affect material degradation in the order black > green > blue > red > orange > white [124]. Carbon black in the black pigment is considered to prevent oxidation [125].
6) Copper conductors may exhibit catalytic effects and promote degradation of insulation. This was confirmed with cables made in the US in the 1970s. At that time their insulation life was estimated to be about 320 years. This catalytic effect is no longer observed for products made after the 1980s [87].
7) EPR subject to accelerated ageing typically shows a sudden drop in EAB [87].
8) Plasticizer vaporization and desorption of hydrochloric acid would be issues for degradation in PVC [61]. Typical degradation behaviour of PVC is that its EAB value temporarily lowers at the beginning and then shows a slight recovery [27], a phenomenon also observed for several other elastomers [13].
9) Elastomers are reported to endure even after 40 years of service if they are not subject to any mechanical stresses [126].
10) Chain scission and cross-linking in butyl rubber [27,127] progresses initially and thereafter periodically.
11) CSPE degrades even in an inert gas. This is caused by a dehydrochlorination reaction [105].
12) Degradation behaviour in neoprene rubber can be explained by oxidation, and the dehydrochlorination reaction can be ignored in this case [94].
13) Pressurized oxygen atmosphere can promote material degradation whilst avoiding inhomogeneous distribution. Details are reported by JAEA [3,45].

5 Accelerated ageing test methods

5.1 Sequential ageing test

5.1.1 General

In general, accelerated ageing tests had been performed as part of the qualification testing requirements of IEEE-323 [128] and IEEE-383 [129] suite of tests, which require type testing of aged and non-aged samples. Historically, the samples were preconditioned by performing thermal ageing followed by radiation ageing. Radiation ageing is a sequence of standard radiation ageing, followed by accident radiation exposure. Table 4 defines this test sequence.

Table 4 – Typical standard acceleration ageing sequence for qualification

Ageing conditon and samples [a]	Aged samples	Unaged samples
Thermal accelerated ageing	Usually at 150 °C	Not aged
Radiation ageing exposure (Cobalt 60 Gamma source) at 5 kGy/h to 10 kGy/h	(Rate 12,5 kGy per year) 0,50 MGy for 40 years 0,75 MGy for 60 years	Not aged
Accident radiation exposure	1,5 MGy	1,50 MGy
[a] Margin not included.		

More recent qualification test sequences carry out radiation ageing first, followed by thermal ageing and subsequently by an accident radiation exposure. This sequence is shown in Table 5.

Table 5 – More recent standard acceleration ageing sequence for qualification

Ageing conditions and samples included [a]	Aged samples	Unaged samples
Radiation ageing exposure (Cobalt 60 Gamma source) at 5 kGy/h to 10 kGy/h	(Rate 12,5 kGy per year) 0,50 MGy for 40 years 0,75 MGy for 60 years	Not aged
Thermal accelerated ageing	Usually at 150 °C	Not aged
Accident radiation exposure	1,50 MGy	1,50 MGy
[a] Margin not included.		

For steam accidents only, such as high energy line break/main steam line break (HELB/MSLB), the conditions are generally better represented by having no accident dose.

5.1.2 Sequence of testing and synergistic effects

Synergistic effects should be considered with respect to the sequence of conducting LOCA simulation tests. One approach is that of US NRC 10CFR50, Part 49 [130]. In that document, preconditioning and testing of equipment must consider known synergistic effects when these effects are known to have a significant effect on equipment performance. The key is knowing which synergies have a functional significance. As environmental requirements become more severe, synergistic effects may become more functionally significant as performance margins are eroded.

This position has been accepted to date, however as technology improves, test sequences and conditions will need to be adapted accordingly. Extensive testing would be required to completely evaluate synergistic effects and their functional significance. Presently, standardized techniques for such evaluation are not available. There is some test evidence which, although it may not cover more recent plant profiles, suggests that there are functional synergies associated with the test sequence as discussed in 5.1.3.

The known technology in this area is limited. For example, a report from the Franklin Institute [131] documents the successful testing of heat shrink tubing using a simultaneous type-test sequence of a) combined thermal and radiation ageing and b) combined radiation and LOCA simulation. SNL, as part of the qualification testing evaluation programme being conducted for the US NRC, has performed similar simultaneous sequence testing on heat shrink tubing and concluded that “no functional synergisms were observed for cable splice assemblies" [132]. Justification for verifying the sequence used in the test programmes mentioned under 5.1.2 as the most severe is based on extensive testing experience of the various cable accessory manufacturers considered state-of-the-art in this area [133].

5.1.3 Guidance on test sequences

IEC 60780 [134] required, and IEC/IEEE 60780-323 [139] requires that the type test and the accelerated ageing tests on equipment shall be run in a specific order. In general, three main test groups should be defined:

Group 1: To check the functional characteristics under normal ambient conditions and in all specified limits of normal operation. IEC/IEEE 60780-323 describes a number of steps necessary to achieve this objective. Briefly, these steps are:

a) equipment inspection;

b) characteristics under normal ambient conditions;

c) tests on the extreme limits of utilization field.

During these different tests at functional limits, the normal ambient condition characteristics determined in test b) (in accordance with IEC 61298-2 [140]) should be checked to see that they are still within the acceptable limits defined in the functional specifications of equipment concerned.

Group 2: To demonstrate the seismic resistance of equipment.

Pre-ageing prior to the seismic test is only required if significant ageing factors exist for the equipment under test.

Group 3: To demonstrate the resistance of the equipment against accident and post-accident conditions. The procedures required are:

1) assessment of behaviour with time;

2) accident and post-accident condition tests.

Since test Group 1 is considered as common industry practice, no additional detailed information is provided. Test Groups 2 and 3 are more specific to nuclear power plants, therefore additional detailed information is provided in 5.1.4. The three test groups may be treated independently and may be applied to different samples of equipment.

5.1.4 Assessment of accelerated ageing

5.1.4.1 General

This step, which generally constitutes the first phase of test Group 3, is of special importance as it ensures that the equipment is ‘aged’ by an accelerated but realistic ageing process before submitting it to accident conditions. Careful consideration of the different tests as well as their sequences (sequential or simultaneous) is required. Some of the key factors required to develop adequate accelerated ageing programmes are discussed in 5.1.4.2 and 5.1.4.3.

5.1.4.2 Ageing concept

Accelerated ageing processes depend on parameters such as activation energy. The ageing process for a specific material or component normally operated at a certain temperature may be simulated by a higher than normal temperature environment or equivalent electrical stresses for a reduced period.

Obtaining reliable accelerated ageing data depends on detailed knowledge of the factors which influence equipment degradation and of the synergies which take place between these quantities. It also requires correct assessment of the dominant ageing processes in any particular case. This means that the selection of ageing tests is non-trivial and that attempts to achieve large acceleration contain a significant risk of error because the ageing mechanism may not be active in the service condition. Accelerated ageing is useful to identify design deficiencies and indicate failure probability, but it cannot completely replace qualification by experience. Not all safety-related equipment is subject to accelerated ageing; in particular, safety equipment which is not supposed to be exposed to accident conditions is not intended to be pre-aged before being seismically tested.

The need for accelerated ageing of particular equipment should be determined on the basis of an evaluation of the specific design and application. At the conclusion of accelerated ageing the test objects are intended to be at the end of life. If previous ageing data for various devices exist, they can be utilized provided these data are applicable and justifiable in regard to the operational conditions that are required by the performance specifications of the device to be type tested. If previous ageing data do not exist, for example in devices comprising different materials, then accelerated ageing is a requirement as part of the test sequence.

5.1.4.3 Distinction between materials and equipment

In tests on materials, only one product is studied at a time. This enables the use of theoretical laws relating to ageing and its acceleration, in cases where they exist, provided the often limited scope of their validity is not exceeded. The testing methods for materials are described in other various normative documents. For electrical insulation, in particular, information will be found in the corresponding IEC publications, and in particular, in IEC 60085 [138].

The situation for equipment is more complex as even a simple equipment is always composed of several different materials. In these situations, the laws and methods of ageing are not only different but sometimes conflicting. Moreover, these materials may give rise to mutual reactions (compatibility) which have their own laws of acceleration. As a result, the accelerated ageing tests carried out on a set of equipment are often very different from those on the various materials of which it is composed.

5.2 Simultaneous ageing test

A simultaneous test combines the various stresses under consideration. Its advantages are a shorter testing time and automatic inclusion of resulting synergies. The disadvantages are the grave difficulties in analysing the results as the ageing stresses are subject to different acceleration laws, and the practical difficulty of building facilities to perform such tests. The sequential method entails subjecting the equipment to a series of tests, each of which involves the application of a single ageing stressor. The advantages are that the specification and interpretation of each test are easier and that a reduced number of standard tests covers a large range of equipment put to identical use. The disadvantages are that only the selected types of ageing are induced, the order in which the tests are carried out becomes very important, and the inclusion of the synergies is complicated (increased severity, selection and order of tests).

A variant of the sequential method consists of at least two stresses in the series of individual tests. In practice, the tests designed to study the ageing of equipment are almost always carried out according to the sequential method or the above variant. It should be noted that whatever the method used (simultaneous or sequential tests), the ultimate goal of environmental qualification testing is to demonstrate, with an acceptable level of conservatism, the reliable operation of equipment under specific operating conditions.

6 Conclusion

This document provides an overview of the influence of temperature on radiation induced degradation of electrical insulating materials, with a focus on most common cable and wire insulations used in nuclear power plants, namely XLPE, FR-XLPE, EPR, FR-EPR, SIR, and PVC. This document, which summarizes and updates the results of CIGRE WG D1.42, offers a historical perspective of insulation degradation research up to the most recent developments and discusses the implications of such research for accelerated ageing test methods.

The data produced over the years by various research laboratories highlights the complexity of the ageing mechanisms under combined radiation and thermal exposure. While this research was able to identify the underlying processes responsible for the observed ageing, attempts to develop mathematical models to explain such processes have only been partially successful. Cable samples harvested from nuclear power plants were found to exhibit limited signs of ageing after 30 to 40 years in service, thus deviating from the original predictions for their lifetime.

Reliable accelerated ageing procedures are required by the industry to maintain the function of electrical equipment insulating materials over the life of the plant and meet regulatory requirements. Sequential ageing remains the preferred method of accelerated ageing in support of environmental qualification of equipment, as defined in IEEE 323 and IEEE 383, mainly because of practical reasons. However, advances in the understanding of insulation materials degradation have led to modifications of the original procedures and guidelines to take into consideration the effects of simultaneous radiation and thermal ageing experiences in the plants.

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