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# Determination of long-term radiation ageing in polymers

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

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# National foreword

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A list of organizations represented on this committee can be obtained on request to its secretary.

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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

INTERNATIONAL
ELECTROTECHNICAL
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INTERNATIONAL ELECTROTECHNICAL COMMISSION

# DETERMINATION OF LONG-TERM RADIATION AGEING IN POLYMERS –

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

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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.

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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- withdrawn,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.

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# 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).

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# 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

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### 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 |

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| 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.

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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].

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