IEC TR 61244-4

Edition 1.0 2019-09

TECHNICAL REPORT

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

IEC TR 61244-4:2019-09(en)

IEC Central Office
3, rue de Varembé
CH-1211 Geneva 20
Switzerland

Tel.: +41 22 919 02 11
info@iec.ch
www.iec.ch

About the IEC
The International Electrotechnical Commission (IEC) is the leading global organization that prepares and publishes International Standards for all electrical, electronic and related technologies.

About IEC publications
The technical content of IEC publications is kept under constant review by the IEC. Please make sure that you have the latest edition, a corrigendum or an amendment might have been published.

IEC publications search - webstore.iec.ch/advsearchform
The advanced search enables to find IEC publications by a variety of criteria (reference number, text, technical committee,…). It also gives information on projects, replaced and withdrawn publications.

IEC Just Published - webstore.iec.ch/justpublished
Stay up to date on all new IEC publications. Just Published details all new publications released. Available online and once a month by email.

IEC Customer Service Centre - webstore.iec.ch/csc
If you wish to give us your feedback on this publication or need further assistance, please contact the Customer Service Centre: sales@iec.ch.

Electropedia - www.electropedia.org
The world's leading online dictionary on electrotechnology, containing more than 22 000 terminological entries in English and French, with equivalent terms in 16 additional languages. Also known as the International Electrotechnical Vocabulary (IEV) online.

IEC Glossary - std.iec.ch/glossary
67 000 electrotechnical terminology entries in English and French extracted from the Terms and Definitions clause of IEC publications issued since 2002. Some entries have been collected from earlier publications of IEC TC 37, 77, 86 and CISPR.

IEC TR 61244-4

Edition 1.0 2019-09

TECHNICAL REPORT

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

INTERNATIONAL ELECTROTECHNICAL COMMISSION

ICS 17.240; 29.035.01

ISBN 978-2-8322-7429-3

Warning! Make sure that you obtained this publication from an authorized distributor.

CONTENTS

INTERNATIONAL ELECTROTECHNICAL COMMISSION

_____

DETERMINATION OF LONG-TERM RADIATION AGEING IN POLYMERS –

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

FOREWORD

1) The International Electrotechnical Commission (IEC) is a worldwide organization for standardization comprising all national electrotechnical committees (IEC National Committees). The object of IEC is to promote international co-operation on all questions concerning standardization in the electrical and electronic fields. To this end and in addition to other activities, IEC publishes International Standards, Technical Specifications, Technical Reports, Publicly Available Specifications (PAS) and Guides (hereafter referred to as "IEC Publication(s)"). Their preparation is entrusted to technical committees; any IEC National Committee interested in the subject dealt with may participate in this preparatory work. International, governmental and non-governmental organizations liaising with the IEC also participate in this preparation. IEC collaborates closely with the International Organization for Standardization (ISO) in accordance with conditions determined by agreement between the two organizations.
2) The formal decisions or agreements of IEC on technical matters express, as nearly as possible, an international consensus of opinion on the relevant subjects since each technical committee has representation from all interested IEC National Committees.
3) IEC Publications have the form of recommendations for international use and are accepted by IEC National Committees in that sense. While all reasonable efforts are made to ensure that the technical content of IEC Publications is accurate, IEC cannot be held responsible for the way in which they are used or for any misinterpretation by any end user.
4) In order to promote international uniformity, IEC National Committees undertake to apply IEC Publications transparently to the maximum extent possible in their national and regional publications. Any divergence between any IEC Publication and the corresponding national or regional publication shall be clearly indicated in the latter.
5) IEC itself does not provide any attestation of conformity. Independent certification bodies provide conformity assessment services and, in some areas, access to IEC marks of conformity. IEC is not responsible for any services carried out by independent certification bodies.
6) All users should ensure that they have the latest edition of this publication.
7) No liability shall attach to IEC or its directors, employees, servants or agents including individual experts and members of its technical committees and IEC National Committees for any personal injury, property damage or other damage of any nature whatsoever, whether direct or indirect, or for costs (including legal fees) and expenses arising out of the publication, use of, or reliance upon, this IEC Publication or any other IEC Publications.
8) Attention is drawn to the Normative references cited in this publication. Use of the referenced publications is indispensable for the correct application of this publication.
9) Attention is drawn to the possibility that some of the elements of this IEC Publication may be the subject of patent rights. IEC shall not be held responsible for identifying any or all such patent rights.

The main task of IEC technical committees is to prepare International Standards. However, a technical committee may propose the publication of a Technical Report when it has collected data of a different kind from that which is normally published as an International Standard, for example "state of the art".

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

Dose rate Gy/h				
Material	**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.