PD IEC/TS 61244-2:2014

BSI Standards Publication

Determination of long-term radiation ageing in polymers

Part 2: Procedures for predicting ageing at low dose rates

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National foreword

This Published Document is the UK implementation of IEC/TS 61244-2:2014. It supersedes BS 7816-2:1997 which is withdrawn.

The UK participation in its preparation was entrusted to Technical Committee GEL/112, Evaluation and qualification of electrical insulating materials and systems.

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

Published by BSI Standards Limited 2014

ISBN 978 0 580 85144 5
ICS 17.240; 29.035.01

This Published Document was published under the authority of the Standards Policy and Strategy Committee on 31 August 2014.

Amendments/corrigenda issued since publication

Date	Text affected

IEC TS 61244-2

Edition 2.0 2014-08

TECHNICAL SPECIFICATION

SPECIFICATION TECHNIQUE

Determination of long-term radiation ageing in polymers –
Part 2: Procedures for predicting ageing at low dose rates

Détermination du vieillissement à long terme sous rayonnement dans les polymères –
Partie 2: Méthodes pour prédire le vieillissement à faible débit de dose

INTERNATIONAL
ELECTROTECHNICAL
COMMISSION

COMMISSION
ELECTROTECHNIQUE
INTERNATIONALE

PRICE CODE
CODE PRIX V

ICS 17.240; 29.035.01

ISBN 978-2-8322-1828-0

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DETERMINATION OF LONG-TERM RADIATION AGEING IN POLYMERS –

Part 2: Procedures for predicting ageing at low dose rates

FOREWORD

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- the subject is still under technical development or where, for any other reason, there is the future but no immediate possibility of an agreement on an International Standard.

Technical specifications are subject to review within three years of publication to decide whether they can be transformed into International Standards.

IEC TS 61244-2, which is a technical specification, has been prepared by IEC technical committee 112: Evaluation and qualification of electrical insulating materials and systems.

This second edition cancels and replaces the first edition published in 1996 and constitutes a technical revision.

This edition includes the following significant technical changes with respect to the previous edition:

a) examples and background information moved to annexes;
b) examples updated with more recent references.

The text of this technical specification is based on the following documents:

Enquiry draft	Report on voting
112/288/DTS	112/305/RVC

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

This publication 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 ageing in polymers*, can be found on the IEC website.

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

- transformed into an International standard,
- reconfirmed,
- withdrawn,
- replaced by a revised edition, or
- amended.

DETERMINATION OF LONG-TERM RADIATION AGEING IN POLYMERS –

Part 2: Procedures for predicting ageing at low dose rates

1 Scope

This part of IEC TS 61244, which is a technical specification, applies to procedures for predicting ageing of polymeric materials at low dose rates.

The object is to present three methods which can be used to extrapolate data obtained from high dose rate experiments to the low dose rates typical of service conditions. These methods assume that homogeneous oxidation has been achieved under the test conditions. The techniques described in the following clauses are methods which have been found to be useful for a range of elastomeric, thermoplastic and thermoset materials. The procedures require a considerable number of test data to enable predictions to be made under low dose rate conditions.

2 Normative references

The following documents, in whole or in part, are normatively referenced in this document and are indispensable for its application. For dated references, only the edition cited applies. For undated references, the latest edition of the referenced document (including any amendments) applies.

IEC 60544-2, *Guide for determining the effects of ionizing radiation on insulating materials – Part 2: Procedures for irradiation and test*

IEC 61244-1, *Determination of long-term radiation ageing in polymers – Part 1: Techniques for monitoring diffusion-limited oxidation*

3 General

The general guidelines of IEC 60544-2 shall be used in the selection of specimen types, radiation source, dosimetry and temperature control. All irradiations shall be carried out in air or at constant oxygen overpressure, although as noted in IEC 61244-1, oxygen overpressure techniques entail some risk of over-ageing the samples. The homogeneity of oxidation through the specimen thickness can be checked using profiling techniques such as those described in IEC 61244-1. The test report shall include details of the irradiation source, dose rate, atmosphere, temperature, sample type and thickness.

All of the procedures described require extensive data obtained over considerable periods of time. Each method has been found to be of practical use within its limitations, but none of the methods can be used where there is more than one mechanism operating with different apparent activation energies.

The power-law extrapolation method (Clause 4) is the simplest of the predictive techniques and requires the least amount of experimental data. This procedure cannot be used at dose rates low enough for thermal ageing to dominate, but appears to be valid for extrapolation of data obtained at near ambient temperatures (20 °C to 30 °C) for polymers such as polyolefins.

Because of the limited data involved, caution should be used in extrapolating by more than a factor of 10 in dose rate.

Both of the superposition methods can make use of data obtained under combined thermal/radiation ageing and are able to predict behaviour in the dose rate regime where thermal degradation is important, but require considerably more experimental data than the power-law extrapolation method. The superposition of time-dependent data (Clause 5) is not applicable to all materials; for instance, it cannot be used with materials which exhibit complex dose rate effects. Where it is applicable, the procedure does lend itself to calculation of the effects of quite complex temperature-dose rate conditions. The superposition of dose to equivalent damage (DED) data (Clause 6) can be used for most materials but, like all of the procedures, it cannot be used to extrapolate through thermal transitions of the polymer.

The general behaviour of polymeric materials aged in radiation environments is described in Annex A.

4 Power law extrapolation method

4.1 Description

This method is based on the extrapolation of radiation ageing data obtained under isothermal conditions in air or in oxygen overpressure over a range of dose rates. The upper limit to the dose rate is such that homogeneous oxidation conditions are achieved. The test data obtained at the different dose rates are used to determine endpoint criteria which are extrapolated graphically to the service dose rate.

4.2 Test procedure

The maximum dose rate at which homogeneous oxidation will occur in the test material shall be assessed. Information in the literature can be used to support an estimation of the maximum dose rate, or to calculate or measure the oxidation layer thickness (IEC 61244-1). Once the maximum dose rate has been established, at least two (preferably three) other dose rates shall be selected, such that the dose rate range covers at least one order of magnitude.

For each of the dose rates selected, samples of the polymer shall be exposed to radiation for at least four ageing times and a property measured that is sensitive to the degradation of the material.

NOTE For cable insulation materials, the measured property would usually be elongation at break; for seal materials, compression set would be appropriate. Suggested properties for other types of component are given in IEC 60544-2.

4.3 Determination of model parameters

The measured damage parameter is plotted against absorbed dose to establish the endpoint at each dose rate. A number of endpoint criteria can be interpolated from the graph (Figure 1); typical endpoint criteria can be the reduction of elongation at break *e* to 100 % or 50 % absolute. A sufficient number of absorbed doses shall be used to enable the endpoint criterion to be established without extrapolation.

The dose at which the end point criterion is reached, i.e. dose to equivalent damage (DED), is plotted against the dose rate in a log-log plot (Figure 2). For most polymers in the radiation-dominated region, this plot is found to be linear, with a slope of *n*, enabling extrapolation to lower dose rates [1][1]. The endpoint dose is then given by

$$\mathrm{DED} = K \cdot \dot{D}^{n} \qquad (1)$$

where

---

[1] Numbers in square brackets refer to the Bibliography.

$\dot{D}$ is the dose rate;

K and n are empirical constants specific to the material tested. The value of the parameter n = <1 and is usually in the range 0 to 0,4.

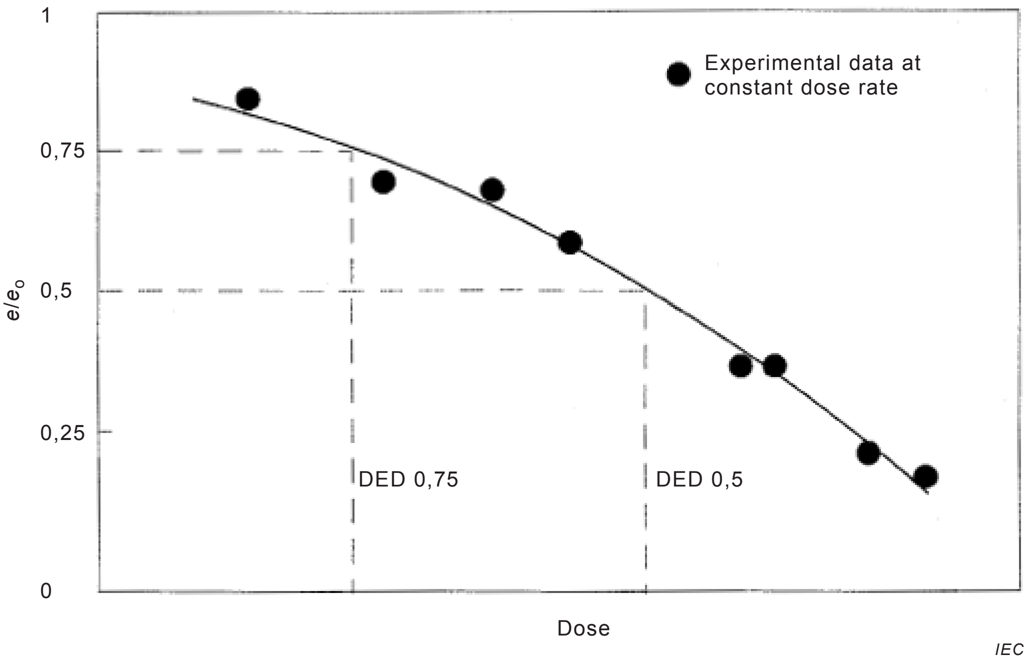
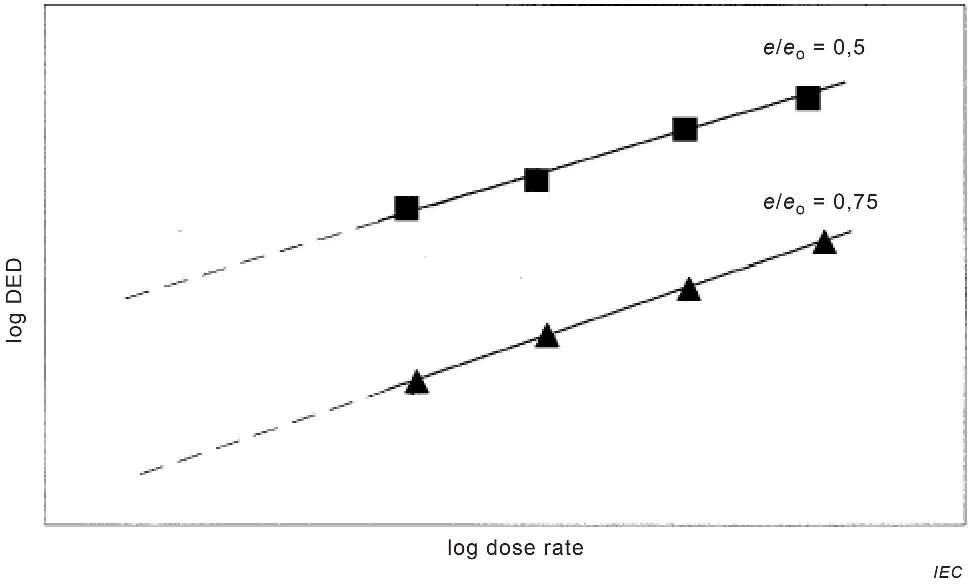


Figure 1 – Interpolation of the end-point dose (schematic), showing a plot of relative elongation at break plotted vs dose with interpolation of DED values at 0,75 and 0,5

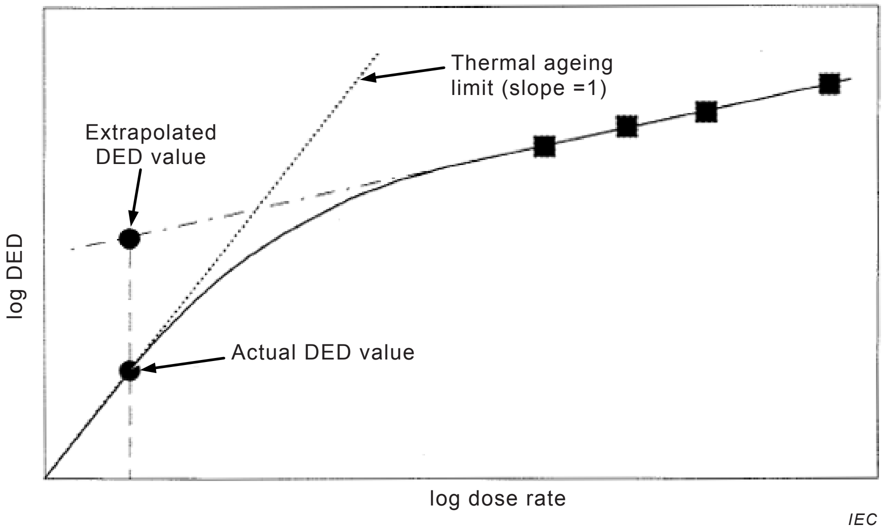


NOTE The slope of the plot for each end-point criterion is the parameter n.

Figure 2 – Extrapolation of end-point dose to lower dose rates (schematic) showing the plot of DED values vs dose rate

4.4 Limitations

This procedure can be a useful method for estimating the behaviour of some polymers at low dose rates but reference to Figure A.1 immediately shows its potential limitations. For all materials they have to break down at dose rates low enough for thermal ageing to become dominant (Figure 3). On a log-log plot of DED versus dose rate used for extrapolation, the thermal ageing limit is represented by a line of slope = 1, i.e. constant time conditions, whereas the slope of the extrapolated data is generally < 1. Extrapolation to dose rates within the thermally dominated region would give unrealistically high values for the predicted DED. This problem can be partially accounted for if separate thermal ageing data are available; these would allow determination of the appropriate thermal only result. If the additional data indicate that thermal effects will dominate, the thermal results can be used for predictions. The power law extrapolation method also cannot be used for materials which exhibit complex dose rate effects such that the log-log plot of DED versus dose rate is non-linear.



NOTE Extrapolation using the parameter *n* will give significantly higher estimates of DED if extrapolations are made near to the thermal ageing limit.

Figure 3 – Limitations – Extrapolation of DED near thermal ageing limit (schematic)

Although the linear extrapolation method assumes that homogeneous oxidation conditions have been obtained in all of the experiments, it appears to be useful in some materials at dose rates where heterogeneous oxidation would be expected to occur. This may arise because cracks generated in the thin oxidized surface layer can then propagate through the bulk unoxidized material, so that the observed macroscopic properties are determined by degradation in that surface layer.

Some examples of the use of the power law extrapolation method are given in Annex B.

5 Superposition of time dependent data

5.1 Description

The second procedure which can be used to extrapolate to lower dose rates makes use of additional data obtained at elevated temperatures under irradiation. The method uses the superposition principle which has been used extensively for thermal ageing (time-temperature superposition). In this method it is extended to time-temperature-dose rate superposition for combined thermal-radiation environments [2, 3]. The basic principle of the superposition technique is described in Annex C.

5.2 Test procedure

Data shall be obtained at a minimum of three dose rates and at least two, preferably three, temperatures at each of these dose rates. For each of these temperature-dose rate conditions, measurements shall be made at a minimum of three times. In addition, thermal ageing data on unirradiated material shall be obtained for at least three temperatures. This is the minimum data set for this method; more accurate assessment of the model parameters will be obtained if more data are available.

5.3 Determination of model parameters

The model based on the superposition of time-dependent data is described by the following semi-empirical relationship between the superposition shift factor $a(T, \dot{D})$ and the temperature and dose rate [3]. It has been shown to be useful for a number of polymers:

$$a\,(T,\ \dot{D}) = \exp\{\,-E/R\,(1/T - 1/T_{\mathrm{ref}})\}\ [1 + k\ .\ \dot{D}x\ .\ \exp\{Ex/R\,(1/T - 1/T_{\mathrm{ref}})\}] \qquad (2)$$

where

T is the temperature in Kelvin;

T_{ref} is the reference temperature; i.e. the temperature at which $a\,(T,0) = 1$. For ease of assessment, T_{ref} is usually chosen to be one of the temperatures used in the combined thermal-radiation ageing measurements.

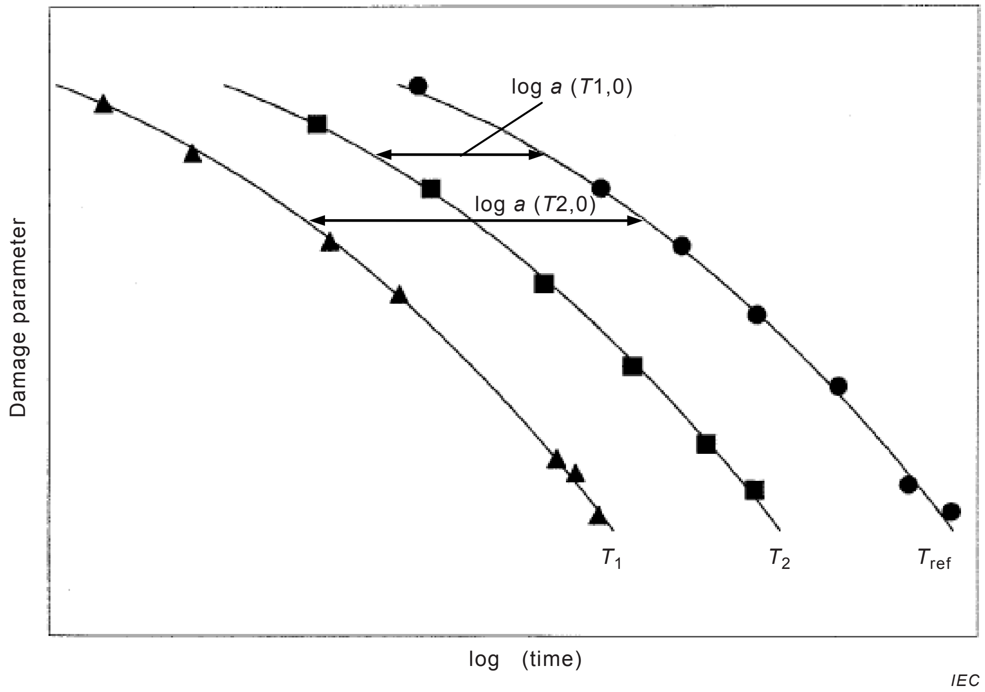
$\dot{D}$ is the dose rate;

R is the gas constant (8,314 $\mathrm{J.mol^{-1}.K^{-1}}$);

E, k and x are the model parameters. The parameter E is the value of the activation energy for thermal-only ageing. The parameters k and x are independent of temperature and dose rate, and determined by fitting the values of $a\,(T,\ \dot{D})$ obtained experimentally to the empirical equation above.

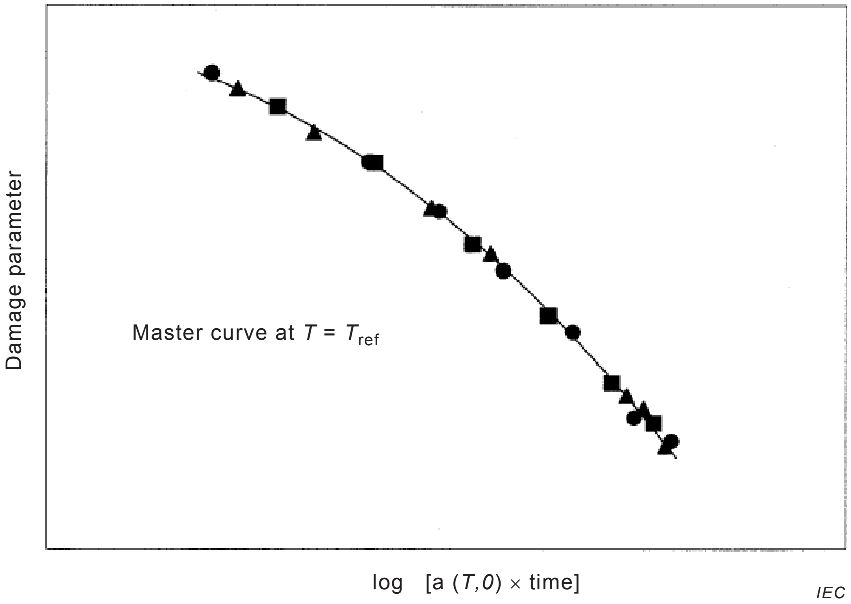
Determination of the model parameters E, k and x is carried out in several stages (as illustrated by Figures 4 to 8).

The first stage in the evaluation is to superpose plots of the damage parameter versus log (time) obtained for thermal-only ageing to yield a master curve (Figures 4 and 5).



NOTE The curves represent measurements of a damage parameter (e.g. elongation at break) as a function of ageing time at three different temperatures, one of which is the reference temperature T_{ref}.

Figure 4 – Determining shift factors a (T,0) for thermal ageing



NOTE The experimental data is superposed using the values of a (T, 0) shown in Figure 4 to form a master curve.

Figure 5 – Superposition of data to yield master curve

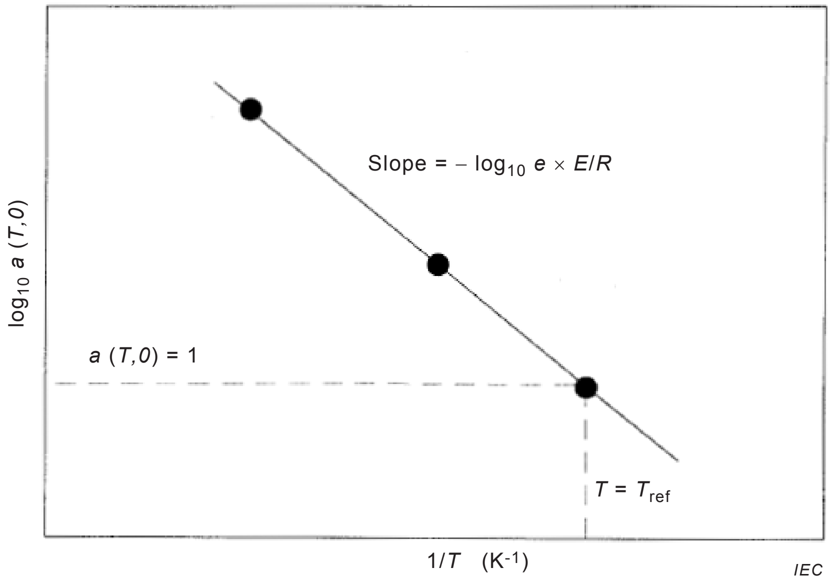
For the condition where $\dot{D} = 0$, Equation (2) simplifies to the Arrhenius relationship:

$$a\,(T,0) = \exp\{-E/R\,(1/T - 1/T_{ref})\} \qquad (3)$$

where

$a\,(T,0)$ is the shift factor for thermal-only ageing.

The values of the shift factor $a\,(T,0)$ required to superpose the data at each temperature can then be plotted versus $1/T$, where T is the temperature in Kelvin (Figure 6). The activation energy E for thermal-only degradation is then determined from the slope of the straight line plot using Equation (3).



NOTE The shift factors a (T, 0) are plotted against $1/T$ to determine the parameter E.

Figure 6 – Determination of activation energy *E*

In the second stage of the evaluation, the time dependent data obtained under combined thermal-radiation ageing conditions are superposed on the master curve as shown in Figure 7. The shift factors $a\ (T, \dot{D})$, at temperature T and dose rate $\dot{D}$, required to superpose the data are determined for each temperature-dose rate condition. At this stage of the evaluation, values of the shift factor $a\ (T, \dot{D})$ are known for the matrix of temperatures and dose rates used.

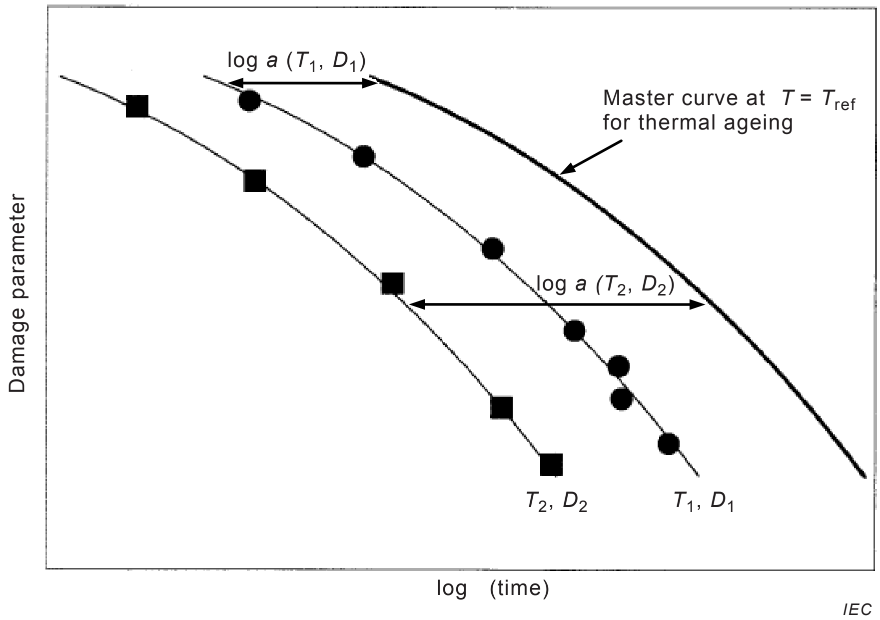


Figure 7 – Determination of shift factors a (T, $\dot{D}$) for combined thermal-radiation ageing, relative to the master curve in Figure 4

The values of $a\ (T, \dot{D})$ are then plotted against the dose rate $\dot{D}$ on a log-log plot (Figure 8). The limiting slope of this plot at high dose rates is the parameter x, since for the condition $T = T_{\mathrm{ref}}$, Equation (2) simplifies to

$$a\ (T_{\mathrm{ref}}, D) = 1 + k \cdot \dot{D}x \tag{4}$$

The parameter x usually takes the value $x \leq 1$. The parameter k determines the position of the curve on the dose rate axis.

Having determined the parameters E, k and x from the experimental data, the empirical model can be used to calculate the DED at lower dose rates or temperatures. This can be determined using the equation:

$$\mathrm{DED} = \dot{D} \cdot t_{\mathrm{m}} / a(T, \dot{D}) \tag{5}$$

where

t_{m}	is the time required to reach the selected damage level at the reference conditions of $T = T_{\mathrm{ref}}$ and $D = 0$ (i.e. on the master curve);
$a(T, \dot{D})$	is calculated from Equation (2). This is shown in Figure 9; the limiting slope of the log-log plot of DED versus dose rate is $(1–x)$ at high dose rates.

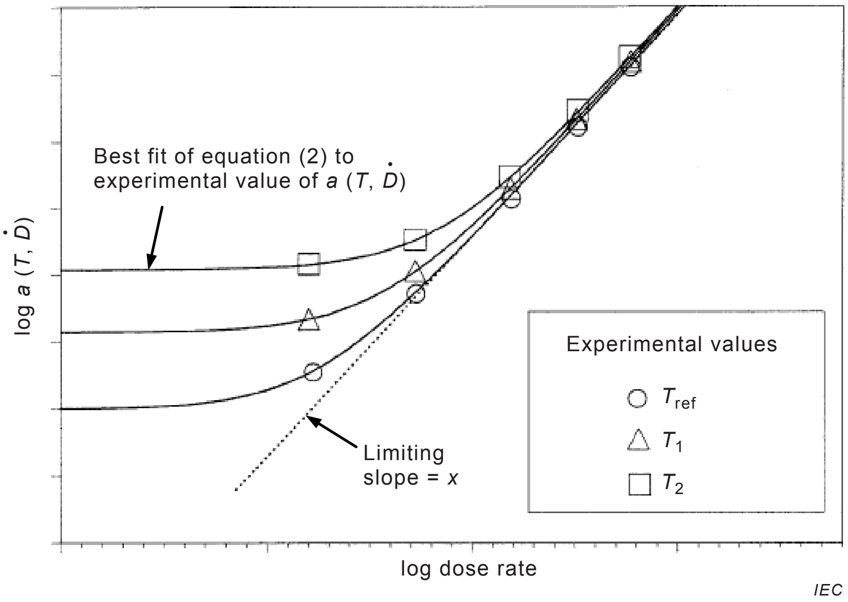


Figure 8 – Fitting experimental values of a (T, $\dot{D}$) to the empirical model Equation (2)

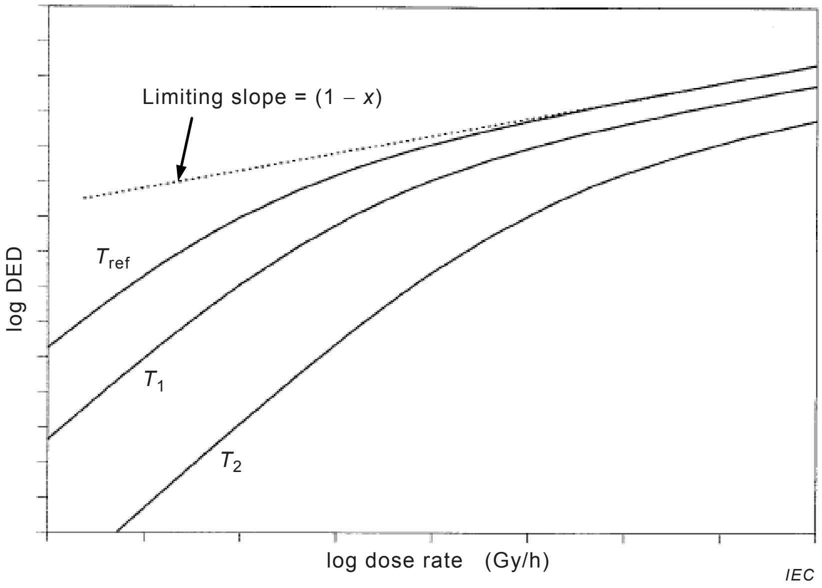


Figure 9 – Calculated DED using Equation (5)

5.4 Limitations

Despite its semi-empirical nature, the general form of the superposition model (Equation (2)) has been found to be of practical use in radiation environments for a range of polymeric components and is particularly useful for elastomeric materials. Some examples of the use of the model are given in Annex D.

This empirical model can only be used for those materials where the shape of the damage parameter versus log (time) curve does not change with temperature and dose rate. In practice, this limits its use to those materials where a single mechanism, e.g. oxidation, dominates both thermal and radiation degradation. If the curve shape changes, superposition of data is not possible and the method cannot be used. The procedure can satisfactorily model the change in DED as the material moves from the radiation dominated region into the thermal dominated region at low dose rate but cannot be used for those materials which show complex dose rate effects. The procedure shall not be used to extrapolate through a thermal transition of the polymer.

6 Superposition of DED data

6.1 Description

This procedure also makes use of data obtained under combined thermal-radiation ageing. Time-temperature-dose rate superposition in this case is applied to plots of log DED versus log (dose rate) rather than damage parameter versus log (time). The shift factors for superposition of DED versus dose rate data are a function of temperature only and are determined by the Arrhenius relationship [4]. This procedure can be applied to a wide range of materials, including those for which superposition of time dependent data is inappropriate.

6.2 Test procedure

Measurements of the damage parameter as a function of dose at several dose rates and at least two temperatures are needed for superposition to be carried out. Sufficient data needs to be obtained at each temperature-dose rate condition for the DED value to be determined without extrapolation. DED can be assessed for a number of damage levels.

6.3 Evaluation

For each temperature-dose rate condition the DED value is determined from a plot of the damage parameter versus dose as shown in Figure 1. These DED values are then plotted versus log (dose rate), noting any data which are subject to DLO effects (Figure 10). The data points are shifted horizontally on the dose rate axis (Figure 11) by using shift factors $a\ (T)$ calculated from the Arrhenius relationship (Equation (6)) with an activation energy equal to that for thermal-only ageing.

$$a\ (T) = \exp \{-E/R\ (1/T - 1/T_{\text{ref}})\} \tag{6}$$

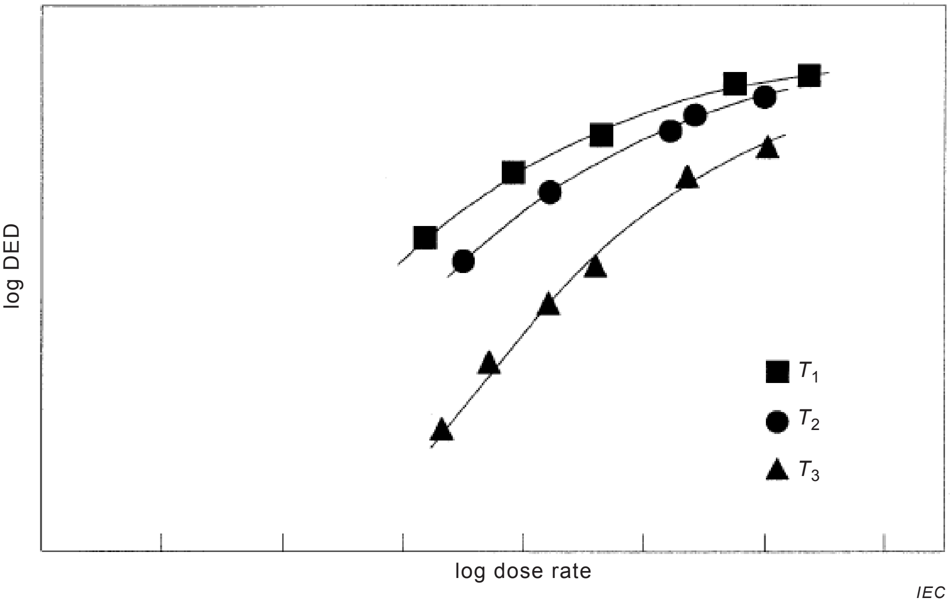
where

E is the activation energy;

T is the temperature in Kelvin;

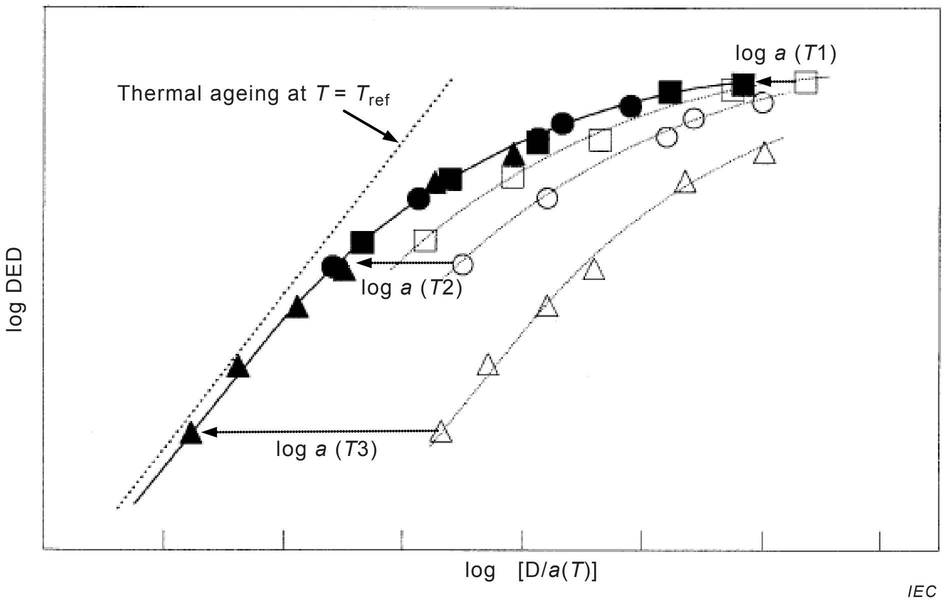
R is the gas constant (8,314 $\text{J.mol}^{-1}\cdot\text{K}^{-1}$);

T_{ref} is the reference temperature.



NOTE DED values for three different temperatures have been determined from plots of the damage parameter versus ageing time, as illustrated in Figure 1.

Figure 10 – DED values under combined thermal-radiation conditions (schematic)



NOTE The DED values shown in Figure 10 have been shifted on the log (dose rate) axis by the shift factors a (*T*), which are determined from thermal-only ageing.

Figure 11 – Superposition of DED data (schematic)

The superposed curve of DED as a function of dose rate enables prediction of the DED at lower dose rates than can be accessed experimentally.

6.4 Limitations

Although this procedure can satisfactorily be used to extrapolate data both in the radiation dominated region and in the thermally dominated region, it cannot be used if the temperature range of interest is at or near a thermal transition of the polymer. The method can also be used for materials that show complex behaviour that cannot be simulated by use of the power law nor the superposition of time dependent data. Some examples of the use of this method are shown in Annex E.

Care shall be taken in using this method for polymeric materials that are semi-crystalline, e.g. crosslinked polyethylene (XLPE), crosslinked polyolefin (XLPO) and some ethylene propylene (EPR). In these materials, reverse temperature effects can occur, such that degradation occurs more rapidly at lower temperatures than at higher temperatures in the presence of radiation. If this effect occurs, DED values plotted as log DED versus log(dose rate) will only superpose for temperature above the crystalline melting point (see Annex E) and the method cannot be used to extrapolate to lower temperatures.

Annex A
(informative)

Behaviour of polymeric materials in radiation environments

Polymeric materials are important in radiation environments because of their use in a range of equipment e.g. as seals and gaskets, cable insulation and jackets. The behaviour of polymers under irradiation is strongly influenced by the atmosphere in which they are irradiated, particularly the presence of oxygen. When polymers are irradiated in oxygen-containing atmospheres, the irradiation dose required to reach a particular level of degradation usually changes with dose rate. The existence of such dose rate effects in the radiation ageing of polymeric materials has been recognised for many years. A sufficient understanding has now been achieved to enable the development of predictive methodologies. The types of dose rate effects which are seen in polymers are illustrated schematically in Figure A.1, where DED is defined as the dose required to reach a specific level of a damage parameter (such as elongation at break, tensile strength, compression set, etc.).

Figure A.1 illustrates behaviour that is seen in most, but not all, polymers. In an inert atmosphere, represented by curve 1, degradation is independent of dose rate over a wide range of dose rates. When dose rates are small enough for thermal degradation effects to dominate, curve I will approach the line representing thermal ageing under inert conditions. On a log-log plot of DED versus dose rate, this thermal ageing will be represented by a straight line of slope equal to one.

In the presence of oxygen, dose rate effects can arise from several processes. At high dose rates, diffusion limited oxidation becomes important (Figure A.1); in this region DED tends to increase with increasing dose rate. It should be noted that Figure A.1 is schematic and is only indicative of the types of behaviour that can occur. The diffusion-limited region in particular is very dependent on the type of polymer, its thickness, the permeation rate for oxygen and the sensitivity of the material to surface properties. The observed degradation is strongly influenced by the thickness of the oxidation layer. At high enough dose rates, oxidation will only occur in a thin surface layer which does not affect the bulk properties of most polymers. In this case, the degradation observed is similar to that seen in an inert atmosphere and DED approaches the inert ageing line. The dose rate above which heterogeneous oxidation occurs can be determined theoretically or by the use of profiling techniques. These procedures are discussed in detail in IEC 61244-1.

In the homogeneous oxidation region, dose rate effects are reduced for many polymers; the slope of the log-log DED versus dose rate plot remains constant or nearly constant with decreasing dose rate (curve 2 in Figure A.1), until the dose rate is sufficiently low for thermal degradation to become dominant. The slope of the DED plot against dose rate is determined by the reaction rate of the dominant chemical reaction. If the reaction rate is high relative to the initiation rate, the slope is small and may approach zero; whereas at low reaction rates, the slope is higher but <1.

In a few polymers irradiated in oxygen-containing environments, more complex dose rate effects are observed in the homogeneous oxidation region.

Additional information on the degradation of polymers can be found in refs [5 – 8]. Much of this information relates to cable insulation materials but the general principles are applicable to other polymeric components.

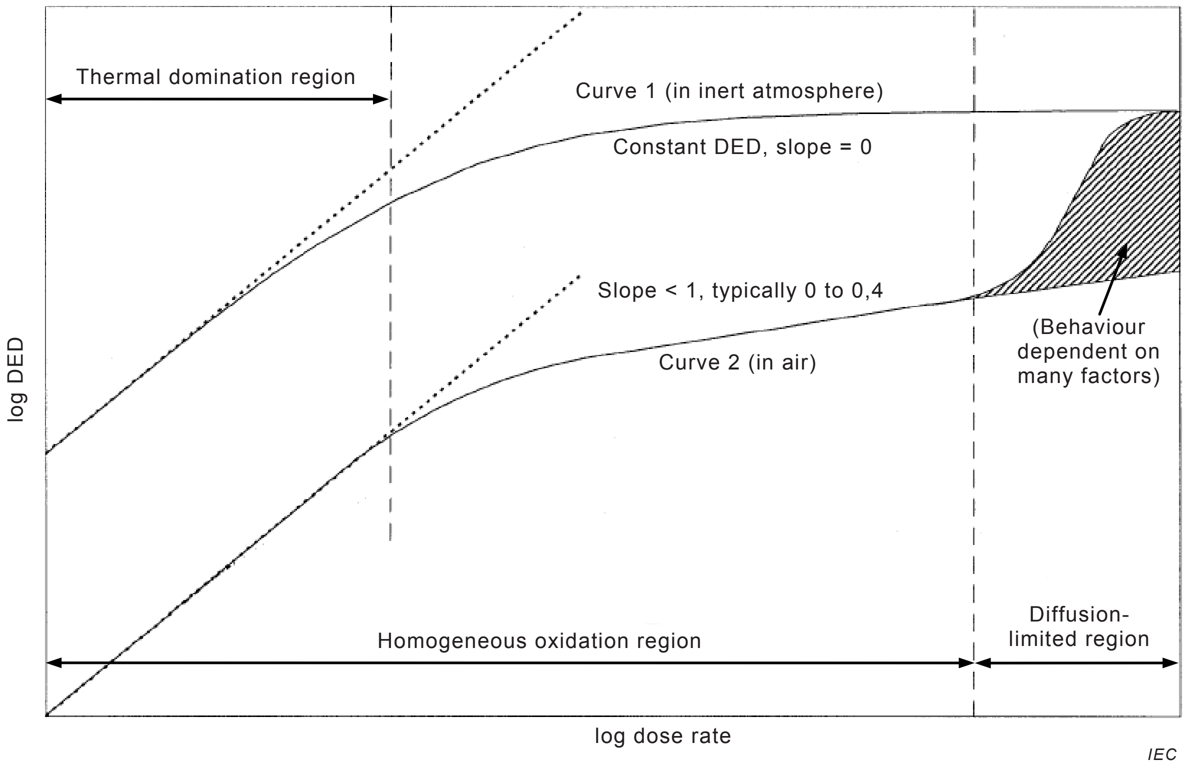


Figure A.1 – Schematic illustrating the types of dose rate effects which can occur in radiation aged polymeric materials

Annex B
(informative)

Examples of use of the power law method

B.1 General

Some examples of the use of the linear extrapolation method are given for several types of polyolefins, as follows.

B.2 Polypropylene filaments

This material was tested over a dose rate range of 4,45 Gy/h to 2 000 Gy/h. In Figure B.1, plots of relative elongation at break e/e_0 are shown for polypropylene monofilaments irradiated in air [1]. Using an end-point criterion of $e/e_0 = 0,5$, the values of the end-point dose DED can be interpolated from these plots. These DED values are shown in Figure B.2; it can be seen that over this dose rate range there is a linear relationship between log (DED) and log (dose rate), with a slope of 0,26.

For this material, homogeneous oxidation was obtained at all dose rates because of the small diameter (0,4 mm) of the samples.

B.3 Crosslinked polyethylene (XLPE)

For this XLPE cable material [9], tested over the dose rate range 9,5 Gy/h to 2,5 kGy/h, data for elongation to reduce to 100 % absolute are shown in Figure B.3. This is an example where heterogeneous oxidation would have occurred at the two highest dose rates. However, for this material, the slope of the plot remains the same at all dose rates, indicating that the elongation is relatively insensitive to the homogeneity of oxidation in this material.

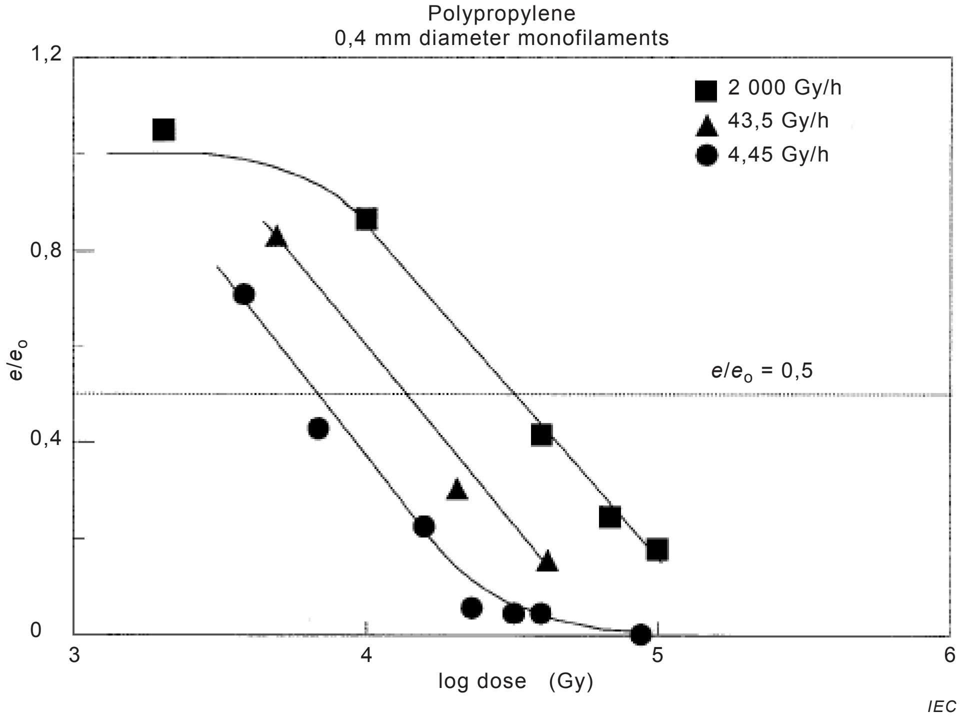


Figure B.1 – Elongation at break of polypropylene irradiated in air (from [10])

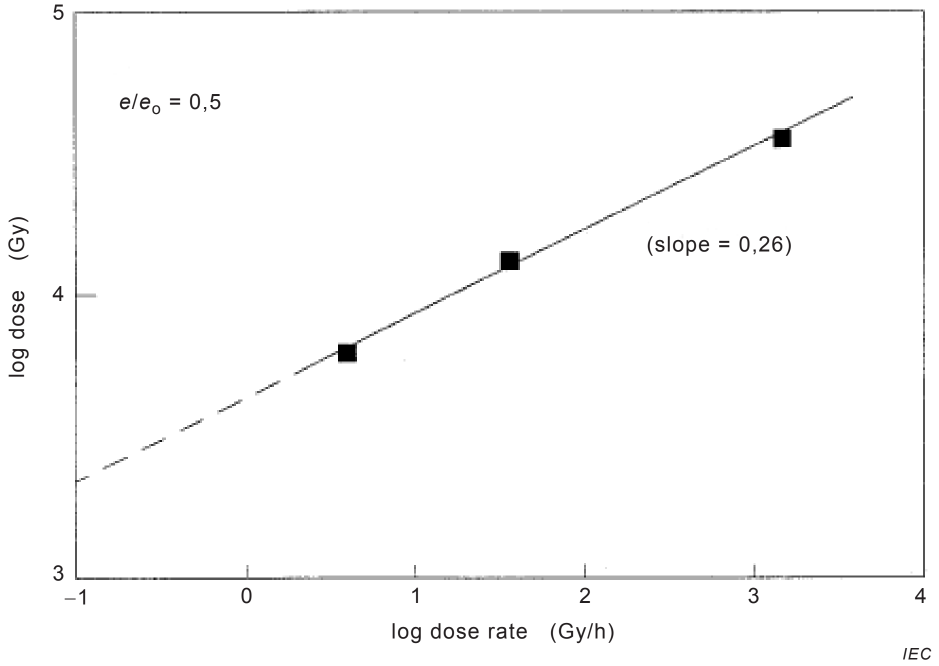


Figure B.2 – Extrapolation of end-point dose from data in Figure B.1

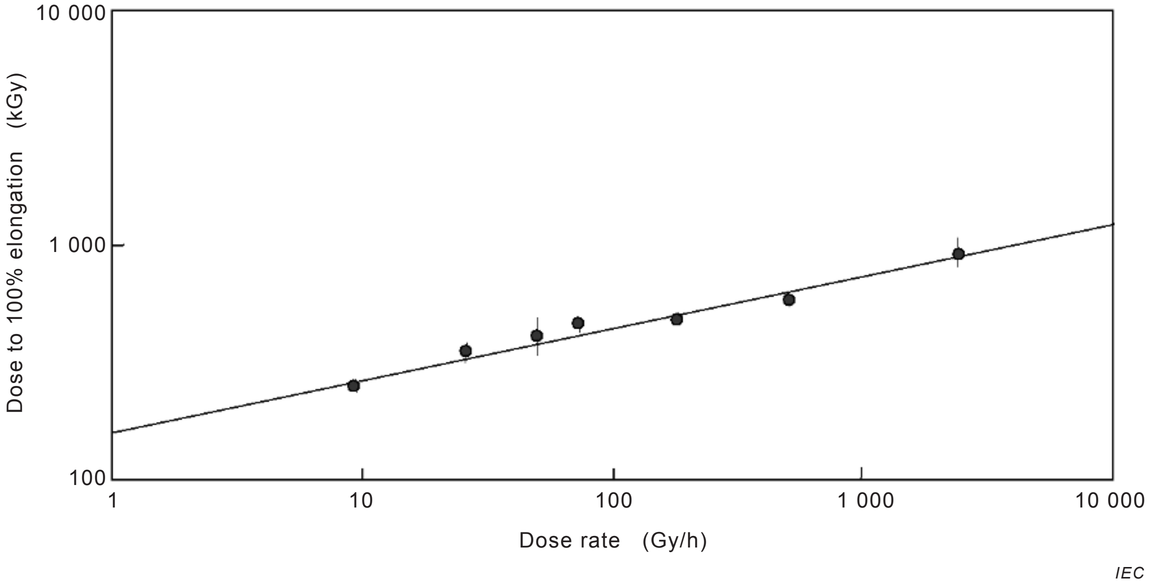


Figure B.3 – Dose required to reach 100 % elongation at 20 °C for an XLPE cable insulation material [11]

Annex C
(informative)

Use of the superposition principle

The superposition procedure in thermal ageing uses time-dependent data taken under isothermal conditions at different temperatures. The higher temperatures are assumed to accelerate the degradation of properties in a uniform manner. Data can be shifted on the time axis to a single curve at the reference temperature by the application of a multiplicative shift factor (Figure C.1). In combined thermal-radiation environments, a similar procedure can be used to shift time dependent data obtained under constant temperature and constant dose rate; this is shown schematically in Figure C.2. By using elevated temperatures to accelerate degradation, data are obtained which are relevant to dose rates lower than those which are accessible experimentally.

In thermal ageing, the functional relationship between the shift factor $a(T)$ and the temperature often takes a simple form, such as the Arrhenius relationship. The empirically derived activation energy from such a functional relationship represents an effective activation energy for the overall thermal degradation.

For combined thermal-radiation ageing, the shift factors $a(T, \dot{D})$ which are determined from the time dependent data do not always show a simple functional relationship with temperature and dose rate. In some cases, an empirical relationship can be determined [2, 3] which enables extrapolation to lower dose rates to be carried out.

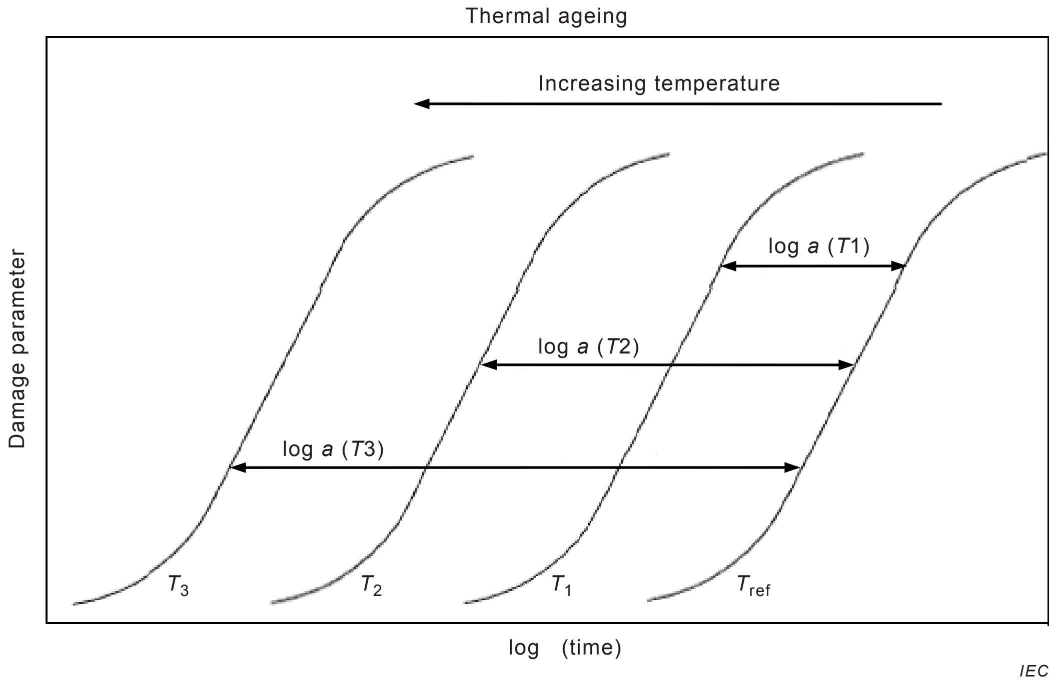


Figure C.1 – Schematic – Superposition principle for thermal ageing

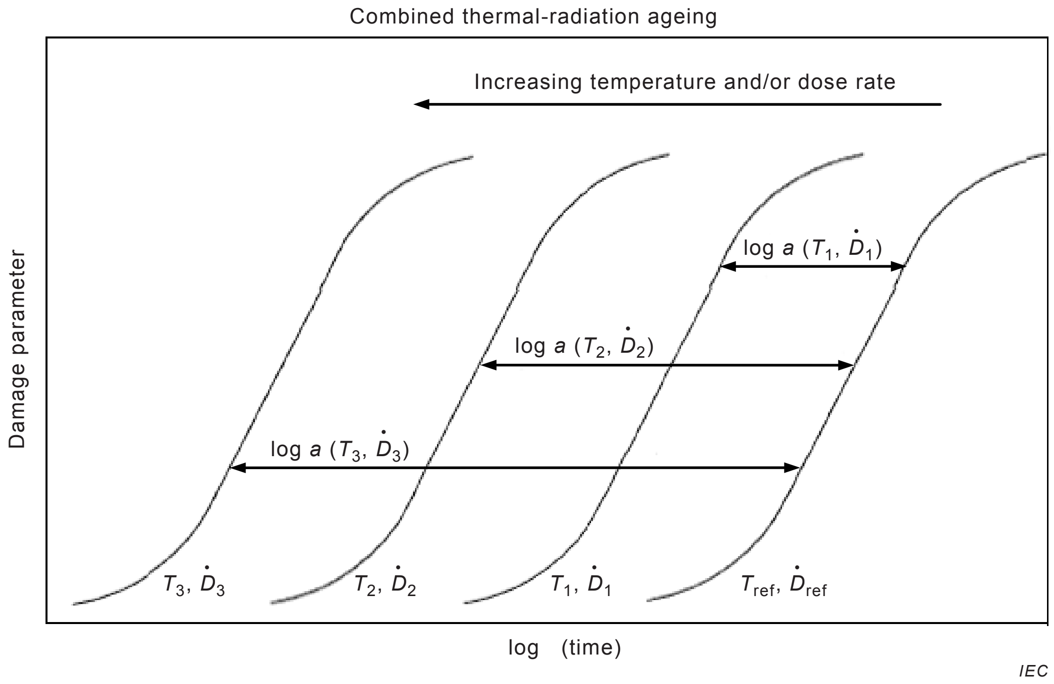


Figure C.2 – Schematic – Superposition principle for combined thermal-radiation ageing

Annex D
(informative)

Examples of use of the superposition of time dependent data

D.1 Ethylene propylene (EPDM) elastomer

This is a formulated elastomer used as a seal material, where compression set has been used as the damage parameter. This material exhibits little temperature dependence (for $T < 90$ °C) under combined thermal-radiation ageing at dose rates in the range 35 Gy/h to 1 000 Gy/h [3]. Using the $a\ (T, \dot{D})$ values obtained in this dose rate range to estimate the model parameters shows that at low dose rates, where thermal effects dominate, the superposition model predicts a marked temperature dependence. Long-term tests at dose rates of 3,5 Gy/h indicate that the model can satisfactorily predict the behaviour of this seal material (Figure D.1).

Using the model to calculate the expected DED values as a function of dose rate shows that, for this material, DED is independent of dose rate over a wide dose rate range. By using data from tests carried out at elevated temperature, the degradation at 20 °C at dose rates down to $1{,}8 \times 10^{-3}$ Gy/h can be assessed (Figure D.2). Note that, in this material, thermal effects dominate at the lowest dose rates illustrating why there is a need for thermal ageing data when using the power law extrapolation method.

D.2 Nitrile elastomer

This material is also used as a seal but, unlike the EPDM elastomer of the previous example, it shows a marked temperature dependence under irradiation at dose rates in the region 100 Gy/h to 1 000 Gy/h [3]. The superposition model has been used to fit the data from this material and can be used to calculate DED values as a function of dose rate (Figure D.3). In this nitrile elastomer, degradation at 40 °C for dose rates <350 Gy/h is dominated by thermal degradation as indicated by the line with a slope of one on the plot of DED versus dose rate in Figure D.3.

D.3 Ethylene vinyl acetate (EVA) polymer

This is a dual layer EVA/EVA cable jacketing material which has been fitted to the empirical model. Unlike the previous two examples where the model parameter x took a value of 1, this material requires an x value of 0,7 to fit the data to the model [10]. A log-log plot of DED versus dose rate then shows a marked dose rate dependence even at high dose rates (Figure D.4); the slope of the plot at high dose rate will be $(1-x)$.

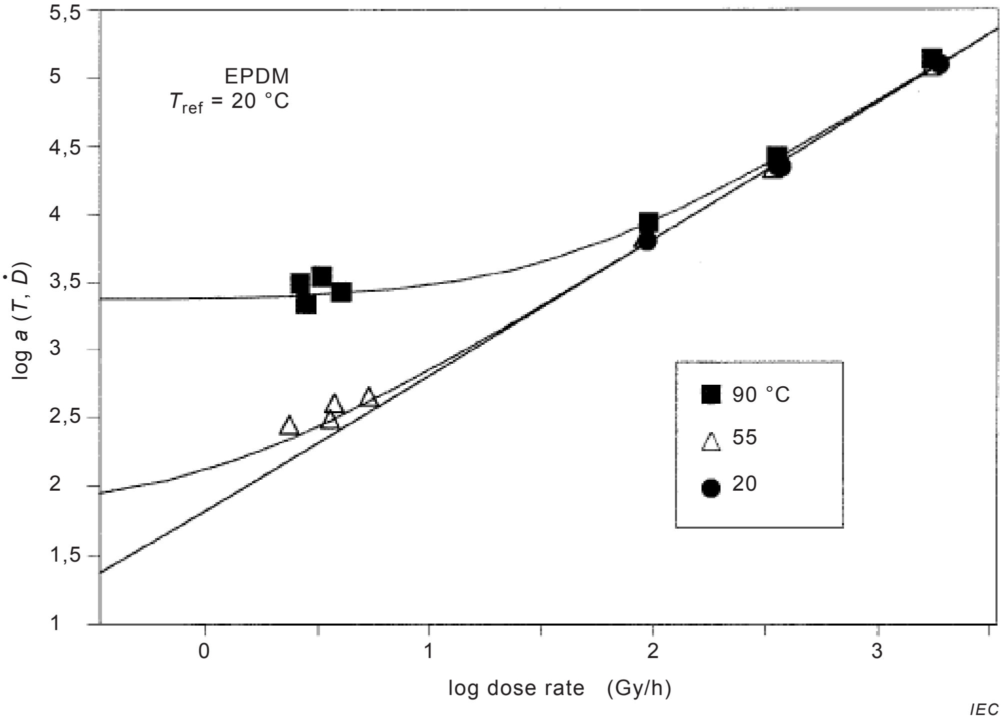


Figure D.1 – Experimental data for EPDM elastomer fitted to the superposition model

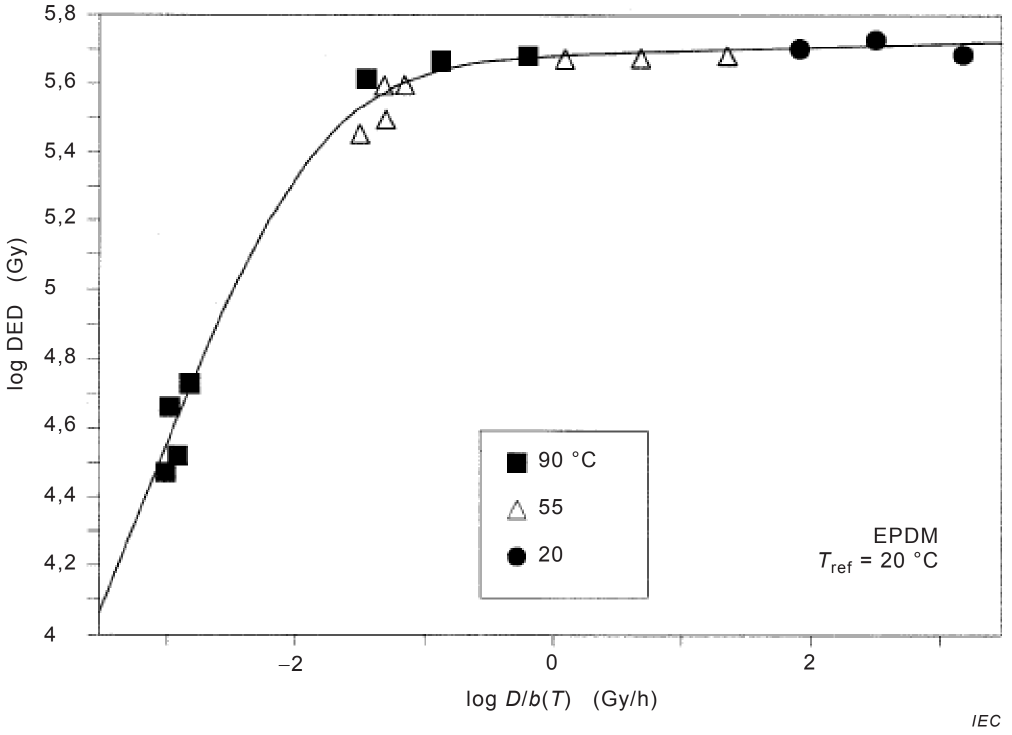


Figure D.2 – Calculated DED for 50 % compression set at 20 °C

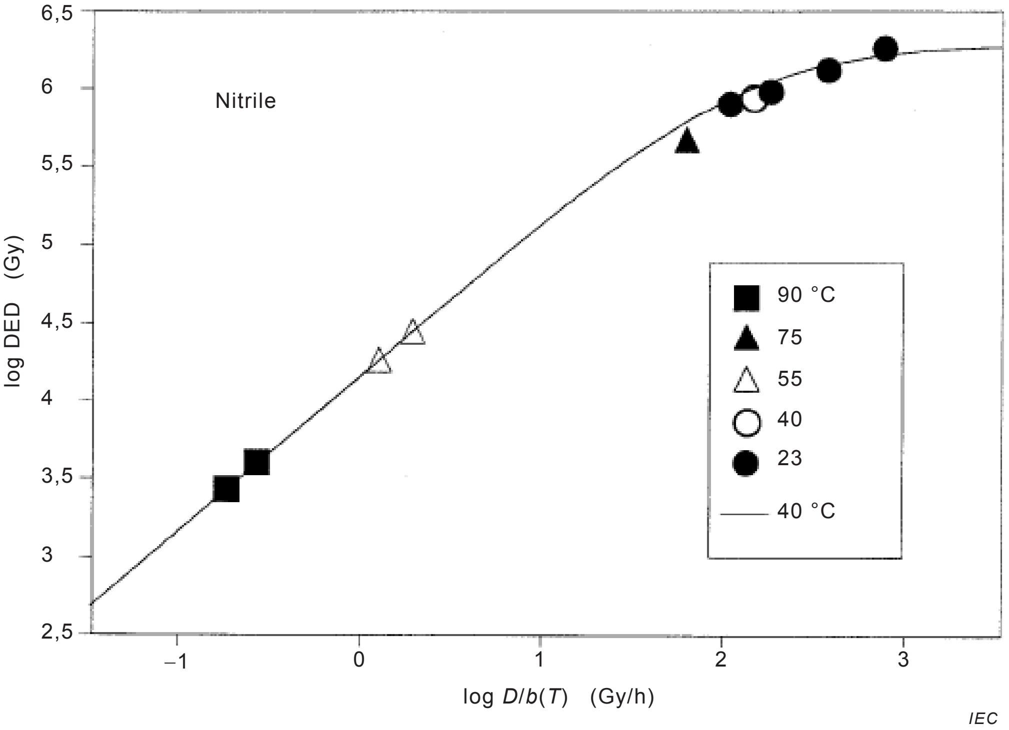


Figure D.3 – Calculated DED for 50 % compression set at 40 °C

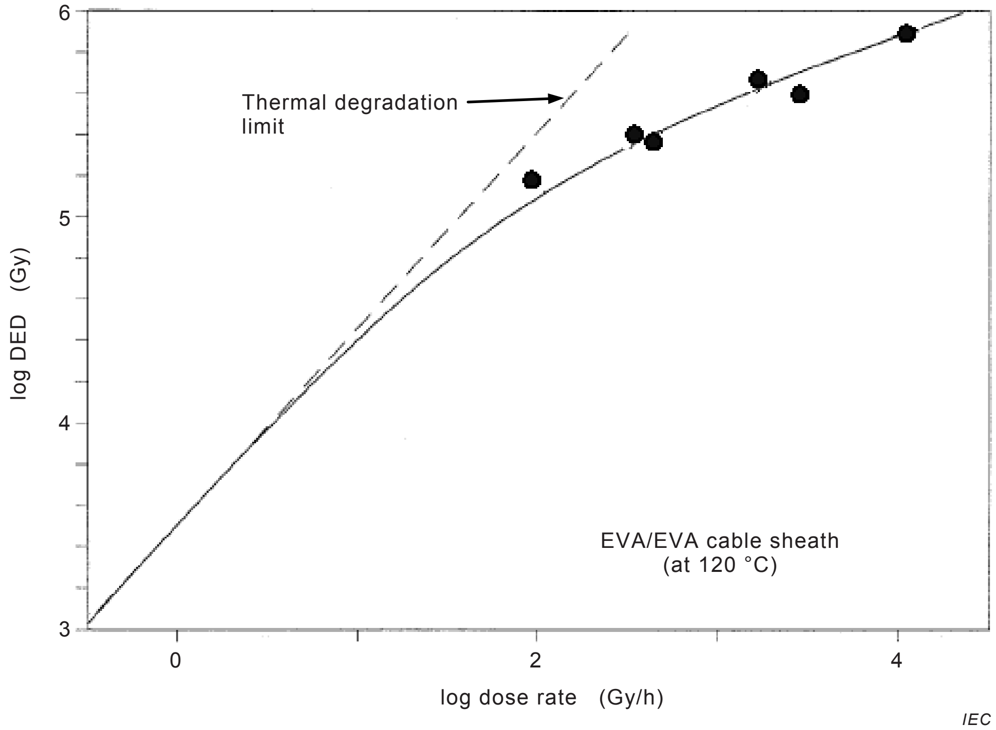


Figure D.4 – Calculated DED for $e/e_0 = 0,5$

Annex E
(informative)

Examples of use of the superposition of dose to equivalent damage (DED) data

E.1 General

Some examples of the use of superposition of DED data are given in the following paragraphs. Numerous other examples are given in reference [12].

E.2 Neoprene cable jacket

Measurements of tensile elongation have been made on this material over a range of combined thermal-radiation environments [7]. Superposition of DED data has been carried out using shift factors determined by the activation energy for thermal ageing. For this particular material, superposition occurs for both homogeneous and heterogeneous data (Figure E.1). It is suggested that this arises because, in heterogeneous samples, cracks which originate in the oxidised surface regions can propagate through the less oxidised bulk material away from the surfaces. This would make the elongation relatively insensitive to the homogeneity of oxidation in the samples.

The superposed data in Figure E.1 can be seen to asymptotically approach the line representing thermal-only degradation, indicating that the thermal environment dominates the degradation in this material at dose rates <1 Gy/h at 50 °C.

E.3 Chlorosulphonated polyethylene (CSPE) cable jacket

Data have been obtained for 6 different CSPE cable jacket materials [7]. All of the data from the different CSPE materials can be superposed on the same curve (Figure E.2), indicating that variations in degradation rate for these materials are relatively small.

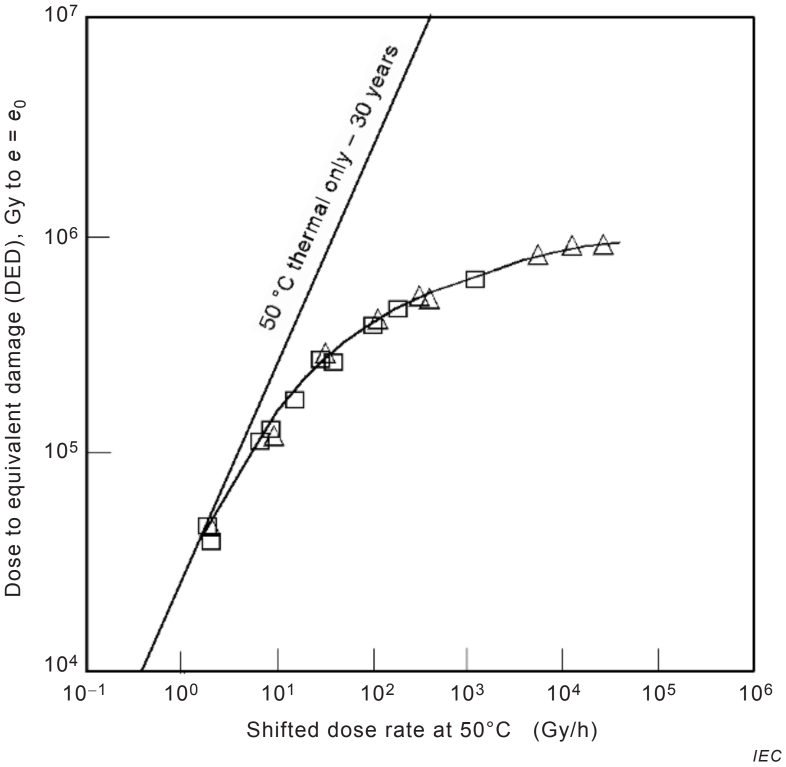
E.4 Crosslinked polyolefin (XLPO) cable insulation

Data on XLPO cable insulation material has been obtained over a range of dose rates from 16 Gy/h to 5 kGy/h over a temperature range of 22 °C to 120 °C [7]. When these data are superposed using shift factors calculated from the activation energy for thermal only ageing, only those data at 60 °C or higher superpose (Figure E.3). At lower temperatures, degradation is much more rapid than expected. This is an example of the reverse temperature effect which is now recognised to occur during radiation ageing of polymers that contain significant degrees of crystallinity. Many polyolefins and some EPR materials are known to exhibit this type of behaviour under combined radiation-thermal ageing conditions [7, 11].

E.5 Poly vinyl chloride (PVC) cable jacket

This particular PVC cable jacketing material has been studied extensively under combined radiation-thermal conditions [4, 13]. An activation energy of 96 kJ/mol yields excellent superposition of the data over a wide range of temperatures and dose rates. Unlike the examples shown in Clauses E.1 and E.2, this material exhibits complex dose rate effects. The superposed data have an unusual S-shaped curve where dose rate effects tend to decrease at low dose rates (Figure E.4).

In contrast to the two previous examples, the activation energy required for superposition does not equal the thermal only activation energy which is 142 kJ/mol. Detailed studies of the kinetics of degradation in this material have shown that the dose rate effects and enhanced degradation at elevated temperatures are due to the rate-determining breakdown of hydroperoxide species produced under irradiation [13, 14] The activation energy for this rate-determining step has been shown to be 96 kJ/mol, i.e. identical to the empirical value found from superposition. The shape of the superposed curve in Figure E.4 is also consistent with the theoretical modelling of the detailed kinetics of the chemical reactions involved.

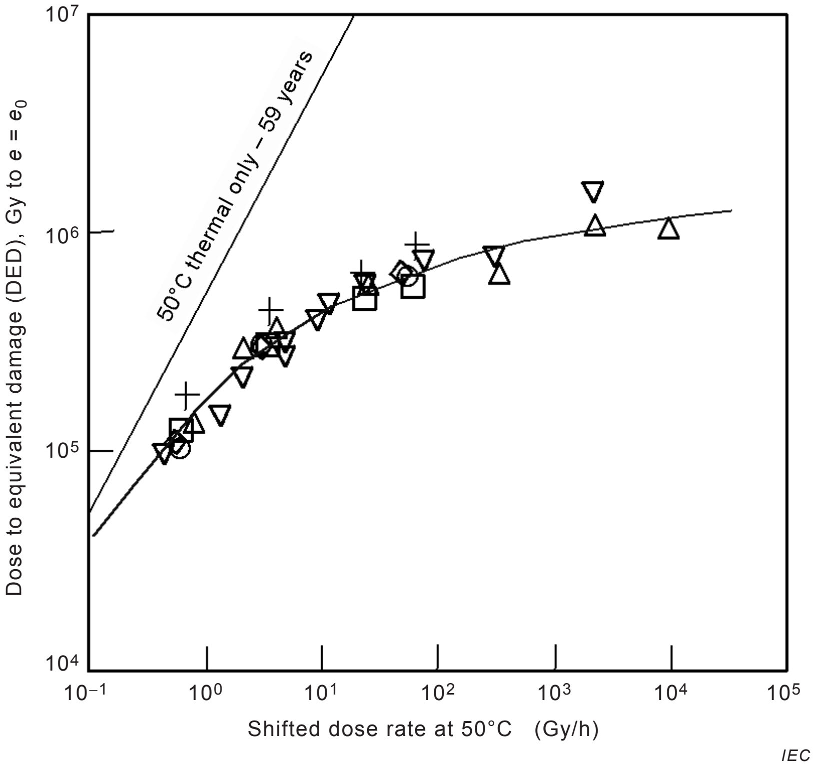


Key

O heterogeneous oxidation

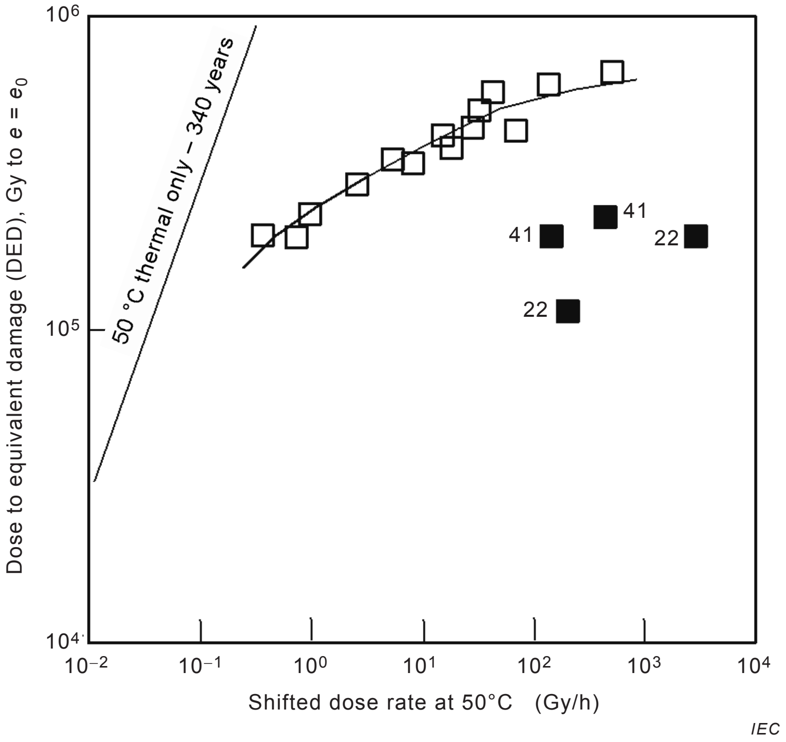
Δ homogeneous oxidation)

Figure E.1 – Superposition of DED data at 50 °C for a neoprene cable jacket material [7]



NOTE Each symbol type refers to a different CSPE formulation [7].

Figure E.2 – Superposition of DED data for several different CSPE cable jacket materials



Key

ϒ radiation ageing data at ≥60 °C

41■ and 22■ data at 41 °C and 22 °C respectively)

Figure E.3 – Superposition of DED data for a XLPO cable insulation material [7]

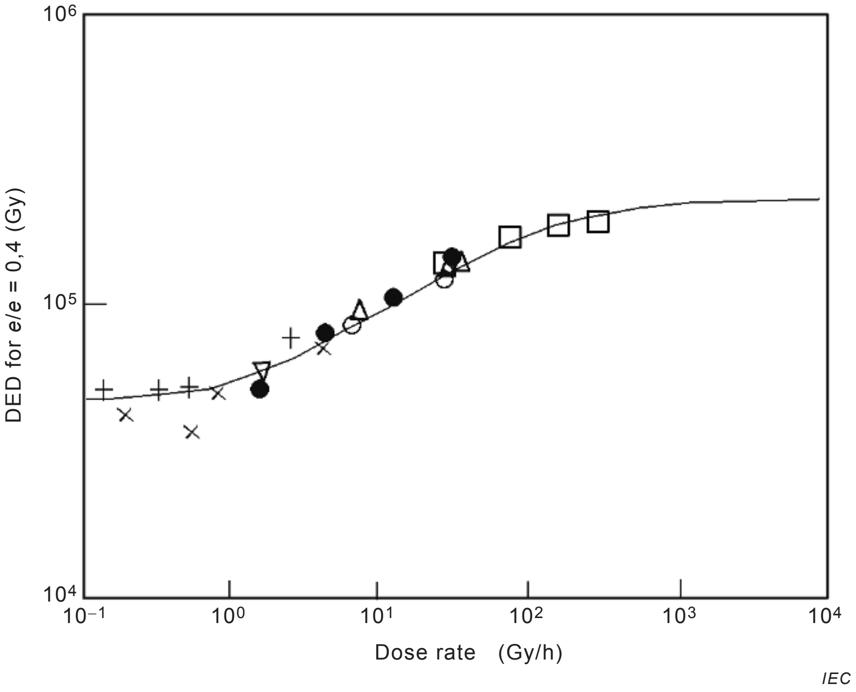


Figure E.4 – Superposition of DED data for PVC showing complex dose rate dependence – Homogeneous oxidation data only

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