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

IEC TS 61244-2:2014-08(en-fr)

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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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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- withdrawn,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.

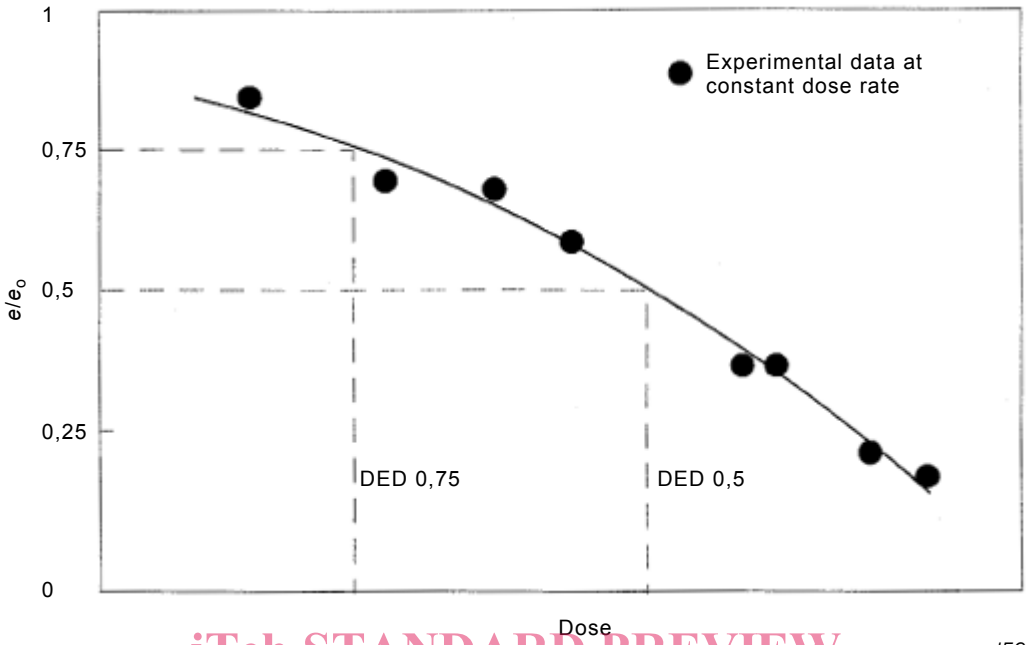
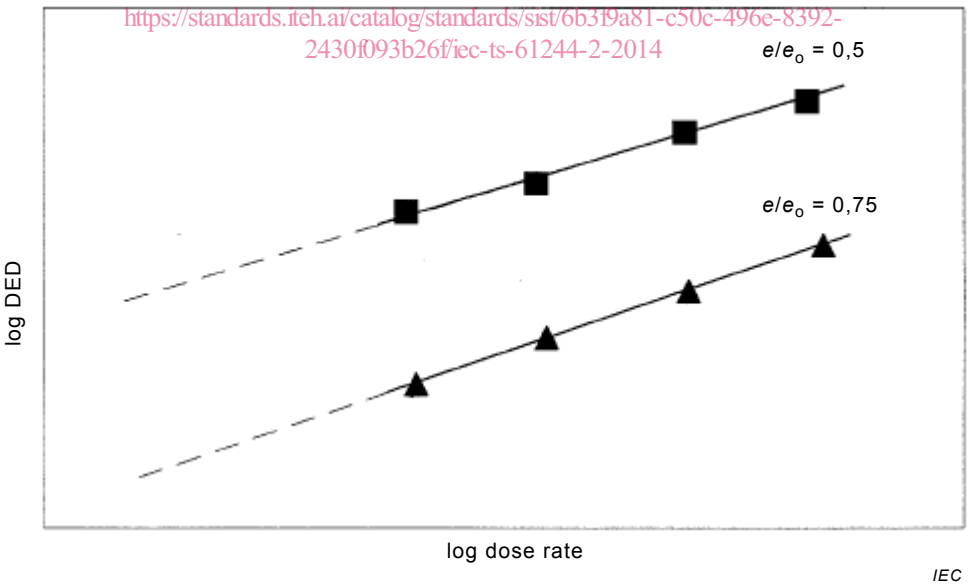


Figure 1 – Interpolation of the end-point dose (schematic), showing a plot of relative elongation at break plotted vs dose with interpolation of DED values at 0,75 and 0,5



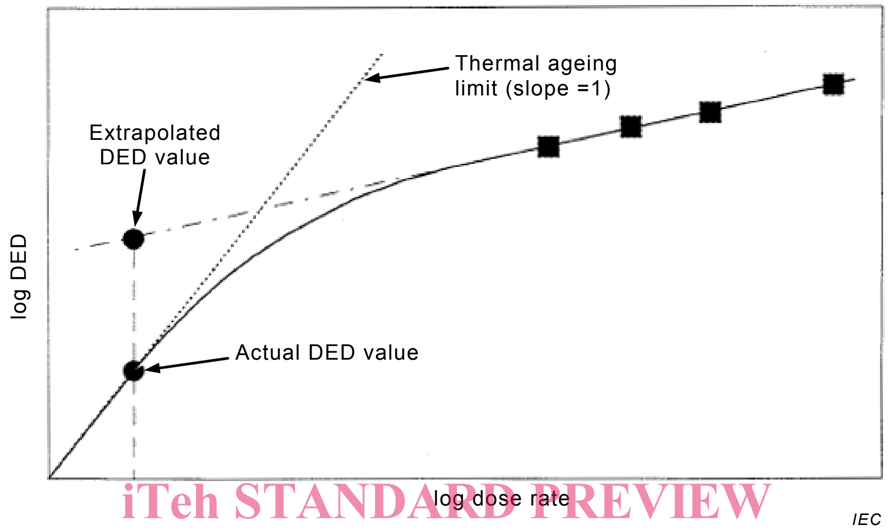
NOTE The slope of the plot for each end-point criterion is the parameter n.

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:

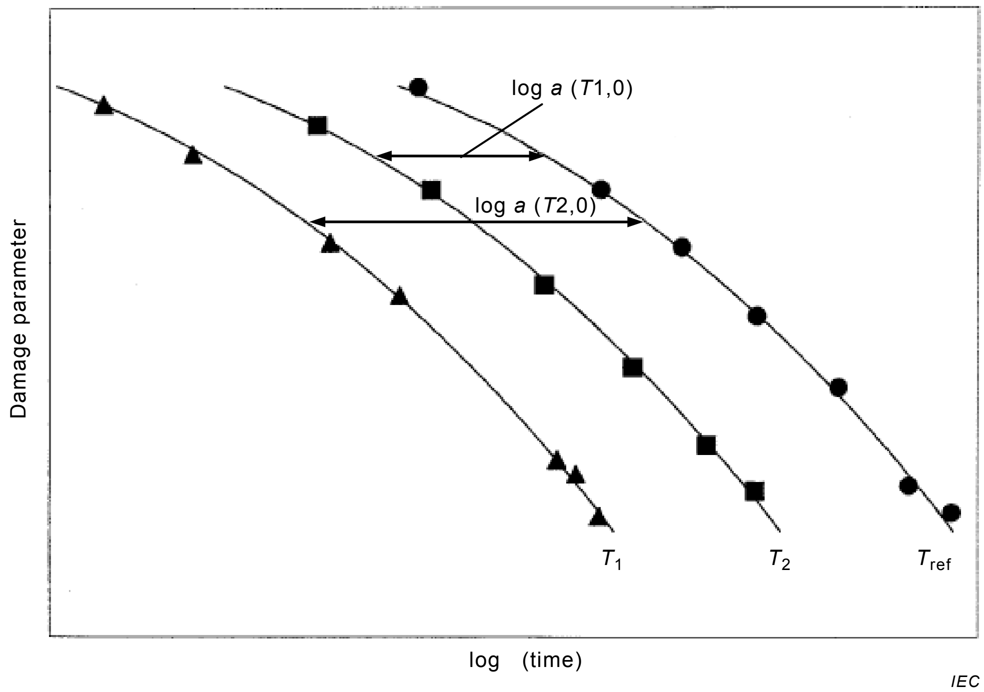
$$a\,(T,\ \dot{D}) = \exp\{-E/R\,(1/T - 1/T_{ref})\}\ [1 + k\ .\ \dot{D}x\ .\ \exp\{Ex/R\,(1/T - 1/T_{ref})\}] \qquad (2)$$

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 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.

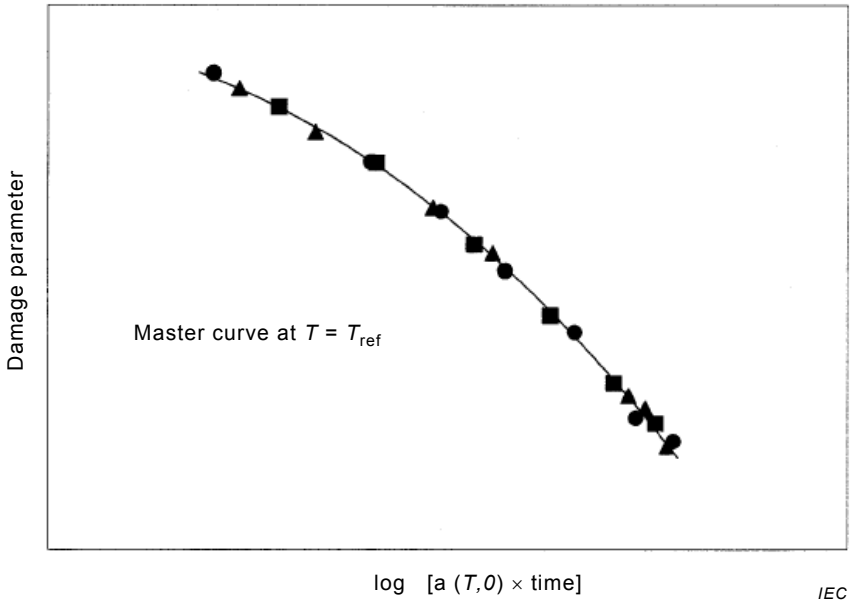
Determination of the model parameters E, k and x is carried out in several stages (as illustrated by Figures 4 to 8).

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

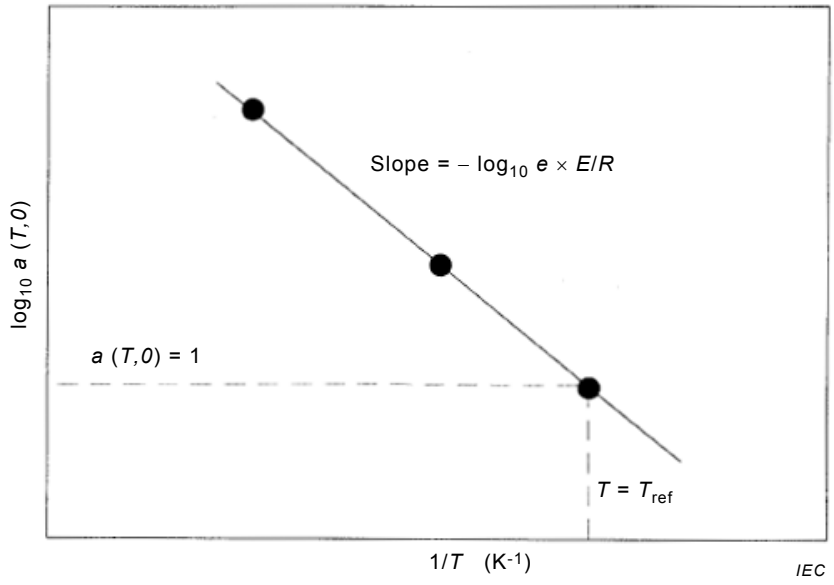
For the condition where $\dot{D} = 0$, Equation (2) simplifies to the Arrhenius relationship:

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

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.

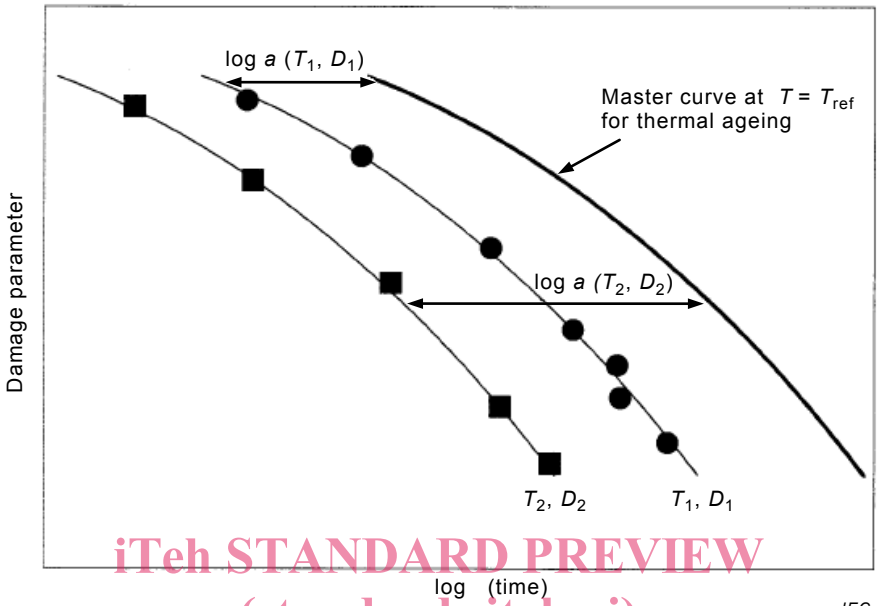


Figure 7 – Determination of shift factors a (T, $\dot{D}$) for combined thermal-radiation ageing, relative to the master curve in Figure 4

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 $T = T_{ref}$, Equation (2) simplifies to

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

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:

$$\text{DED} = \dot{D} \cdot t_m \,/\, a(T, \dot{D}) \tag{5}$$

where

t_m is the time required to reach the selected damage level at the reference conditions of $T = T_{ref}$ 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.

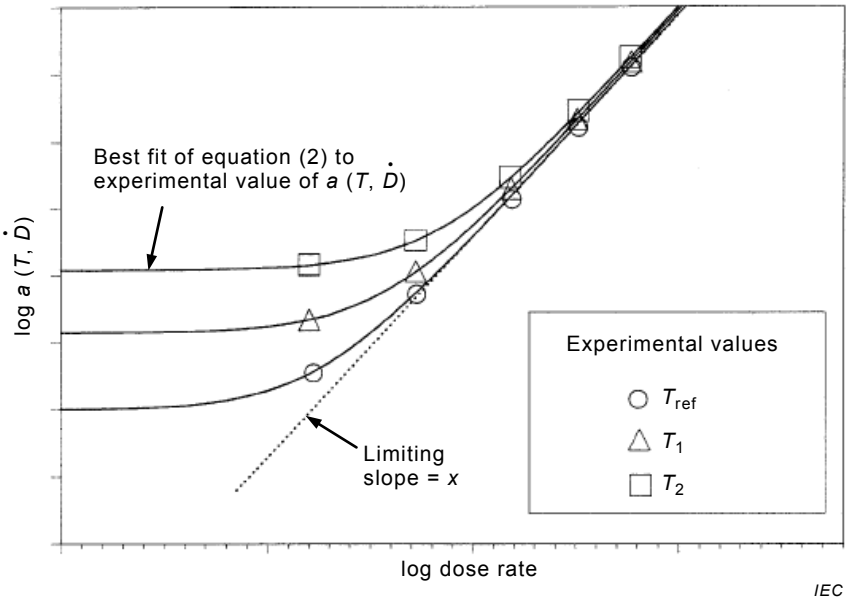


Figure 8 – Fitting experimental values of a (T, $\dot{D}$) to the empirical model Equation (2)

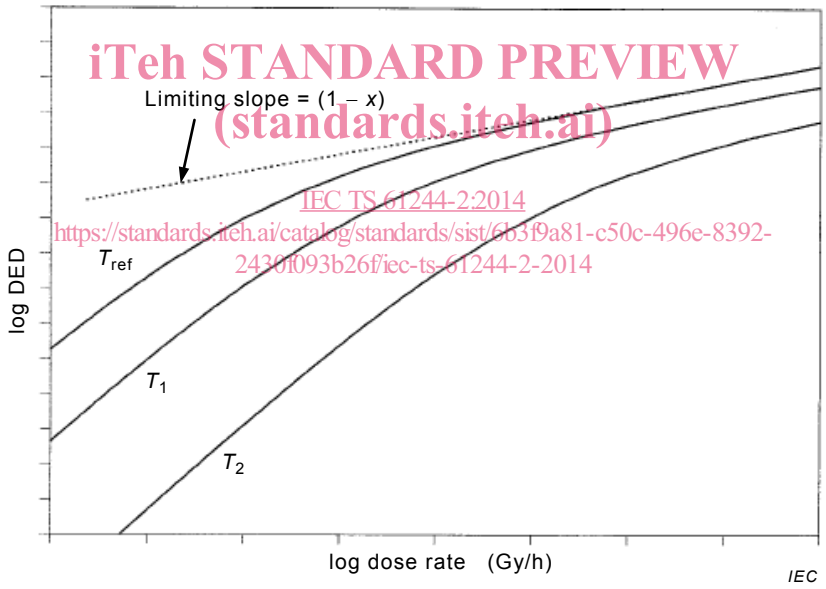


Figure 9 – Calculated DED using Equation (5)

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