Investigation in Thermal Endurance of Polyesterimide Used in Electrical Machines

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ABSTRACT: The purpose of this investigation is to evaluate thermal lifetime of polyesterimide using Weibull statistics. Thermal aging was performed on twisted pair specimens of copper wire insulated by a polyesterimide layer. The study shows that breakdown voltage varies versus aging time. Its increase is allotted to a crosslinking inducing a diminution of mean-free path of charge carriers leading thereby to a mobility decrease, whereas its decrease is attributed to the viscosity diminution expressing a weakening of molecular bonds and a mobility increase of charge carriers. Shape parameter changes in function of aging time. Its increase is ascribed to an arrangement of the molecular structure, whereas its decrease is due to an augmentation in the size of defects. The thermal endurance graph is a straight line indicating that the degradation is governed by a first-order chemical reaction. Activation energy and temperature index were determined. The degradation is governed by the dissolution of copper into the polymer and accelerates its degradation occurring at the polyesterimide–copper interface. Oxygen can diffuse into the insulation and attack copper resulting in the formation of copper oxide. The degradation is caused by the scission of imide and ester bonds. The process is followed by a change in color and a presence of cracks. © 2011 Wiley Periodicals, Inc. J Appl Polym Sci 121: 2886–2892, 2011

Key words: ageing; degradation; failure

INTRODUCTION

A wide variety of solid insulating materials as polyesterimide, polyamide imide, and polyimide are usually used in electrical machines. Their choice is dictated by many factors: working environment, applied voltage, rotating speed, and operating temperature. They are generally characterized by breakdown voltage, capacitance, dielectric loss factor, and flexibility, resistance to abrasion, solvent bonding, mass loss, and temperature index. The thermal class of electrical machines is the main dimensioning factor for solid insulating materials. Polyesterimide is one of the most polymers used in electrical devices as motors, generators, etc. This material has a greater resistance to abrasion and an excellent resistance to high temperature and good electrical and chemical properties.[1] The aging of solid dielectrics is characterized by an irreversible deterioration affecting their performances and their lifetime. The aging phenomena are influenced by the presence of defects, which can be introduced when implementing the material and/or during the manufacturing process of the systems despite all the precautions one can take. Therefore, thermal aging is a very important test in the evaluation of electrical insulation systems.

Thermal aging of polyesterimide has been investigated by many authors.[2,3] Brandes et al.[2] studied thermal aging of polyesterimide at 155, 175, 200, and 220°C. These authors mentioned a decrease of dielectric strength with respect aging time. They reported a change in thickness less than 0.5% after 2500 h at 220°C. They indicated that viscosity of polymer resin over a period nearly 3 years increases with aging time. The Arrhenius curve is a straight line, and the temperature index is 188°C. Araki et al.[3] investigated the degradation of twisted pair samples of polyesterimide aged at 230°C. The results show no significant change of breakdown dielectric strength occurred at the first stage (100 h). However, at the last stage, dielectric strength increases and then decreases abruptly with aging time. This phenomenon is connected to a crosslinking of the material by thermal aging. After this stage, the decomposition of molecular network takes place leading to a diminution of dielectric strength. The degradation is accompanied by a decrease in the thickness of dielectric layer. The degradation of the polymer was examined by thermogravimetric analyzer (TGA), Fourier transform infrared (FTIR), spectrophotometer with a micro-attenuated total reflex ion adaptor, pyrolytic gas chromatograph with FTIR.

The aim of this work is to study the thermal lifetime of polyesterimide. The evolution of breakdown

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voltage in function of thermal aging time has been investigated. The general test procedure in use is the conventional aging according to IEC 216-1.[4] It is well known that the dielectric breakdown of solid insulating materials is a random phenomenon.[5–7] Therefore, the tests were realized on large populations, and the measurement values were analyzed statistically by the two-parameter Weibull distribution.

EXPERIMENTAL

Preparation of samples

The tested samples were prepared as twisted pair specimens of copper wire covered with a 35 μm polyesterimide layer of class H (180°C). A copper wire coil insulated by polyesterimide of grad 2 was supplied by "TREFUCUIVRE" manufacturer in Algeria. The diameter of the wire and the length of the samples are 0.63 and 125 mm, respectively. The specimens were elaborated according to IEC Standard publication 172.[8] To avoid any presence of microscopic cracks, which can constitute a site of partial discharges, the samples were examined under a microscope.

Thermal aging

To estimate the thermal lifetime of polyesterimide, several samples, manufactured as described earlier, were aged in ventilated ovens regulated at different temperatures: 200, 220, 240, 260, and 280°C. After each aging time and for a given temperature, a population of 50 samples was withdrawn and subjected to test of dielectric breakdown.

Test of dielectric breakdown

The two conductors of the samples were connected to a transformer delivering an alternating voltage, which can reach 16 kV, 50 Hz. Breakdown experiments were executed, in air, and at room temperature, by applying voltage increasing with a rate of 0.5 kV/s.

RESULTS AND DISCUSSION

Statistical analysis of breakdown voltage

Among current statistical models, the Weibull model[9] is regarded as the most appropriate for data analysis obtained from breakdown tests of solid dielectrics.[10–12] In the following, this kind of distribution will be used. The cumulative probability P(V), generally used to treat the data of breakdown voltage, is the two-parameter Weibull distribution, which is given by the following equation[13]:

$$P(V) = 1 - \exp\left[-\left(\frac{V}{\alpha}\right)^{\beta}\right] \tag{1}$$

where V is the breakdown voltage, α the scale parameter representing the breakdown voltage for a probability of 63.2%,[14] β the shape parameter which is the slope of the straight line of Weibull plot, with α and V in kV.

The experimental data plot must be a straight line in the coordinate system:

$$X = \log V \tag{2}$$

$$Y = \log\left(\text{In}\frac{1}{1-p}\right) \tag{3}$$

where P is the cumulative breakdown probability.

In this investigation, the measured values of breakdown voltage were plotted according to the two-parameter Weibull distribution. The different steps of this method were presented in a previous work.[15] The data were classified by ascending order, that is, from the smallest to the largest one. The cumulative breakdown probability is calculated for each value V_i using the following relationship[16]:

$$P_i = \frac{i}{N+1}100\% \tag{4}$$

where P_i is the cumulative failure probability in %, N the total number of tested samples, and i the value rank of breakdown voltage V_i. In this work, N was fixed equal to 50.

For the calculation of confidence intervals and curve fit, the maximum likelihood method and the least-squares regression method are usually used.[14] In this study, the maximum likelihood method was chosen, because the confidence intervals calculated by the least-squares regression method are much narrower than those obtained by the first one. Therefore, the use of the least-squares regression is statistically an incorrect method.[17] In addition, it was reported that the maximum likelihood method is generalized to treat data of dielectric breakdown.[18] The best linear fit of Weibull plot and the 90% confidence intervals were determined. Then, the nominal breakdown voltage (63.2%) or the scale parameter and the shape parameter were deduced. Figure 1(a–f) presents some examples of Weibull graphs of breakdown voltage before and after aging at the different temperatures. As can be seen, the width of confidence intervals is more important for the lower probabilities. One observes that the data can be partly or totally inside the tolerance intervals indicated by discontinuous lines. The results of statistical

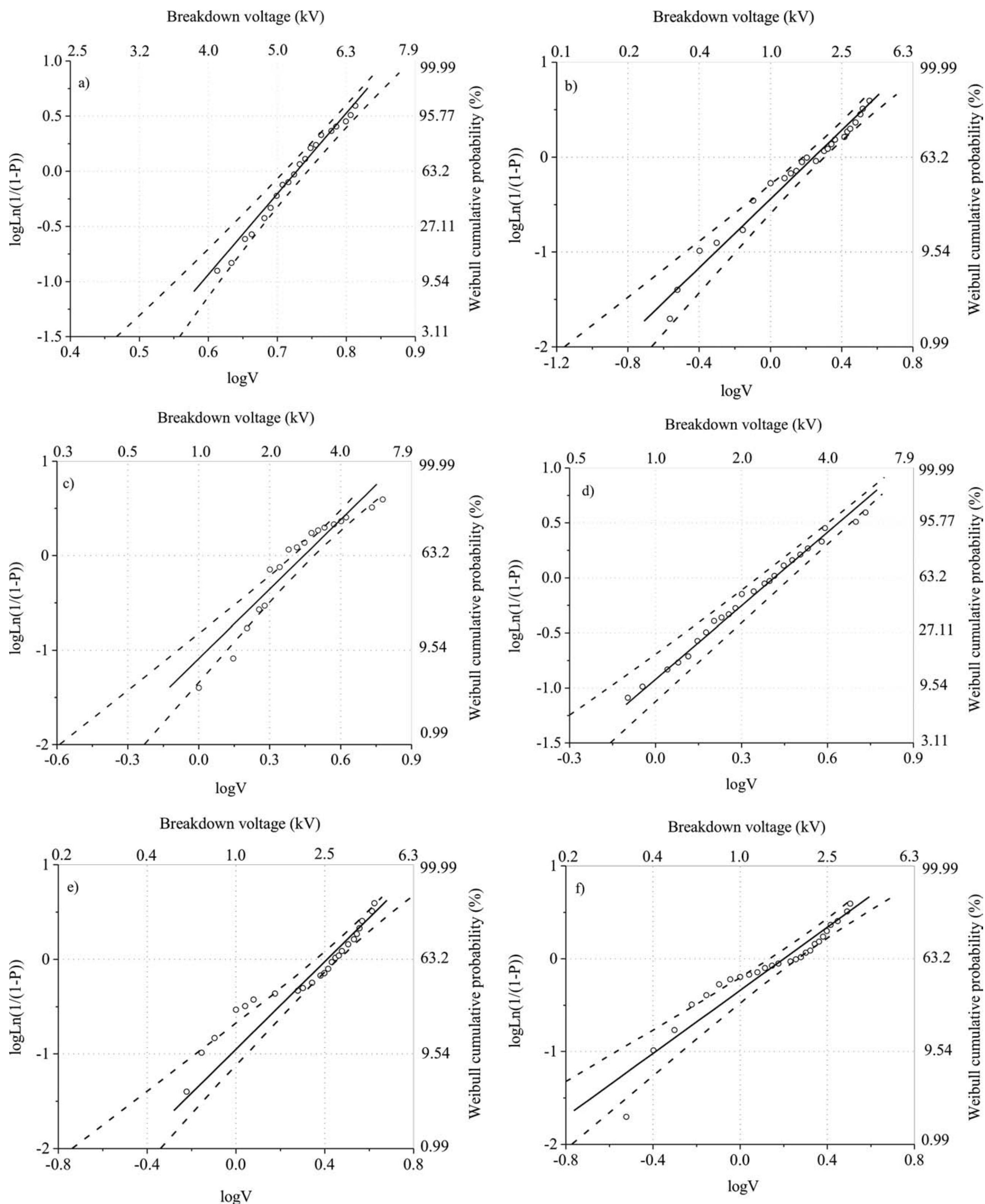

Figure 1 Weibull plot of breakdown voltage data with 90% confidence intervals before and after aging, (a) before aging, (b) after 6000 h at 200°C, (c) after 2700 h at 220°C, (d) after 1300 h at 240°C, (e) after 520 h at 260°C, and (f) after 140 h at 280°C.

analysis were summarized in Table I. The statistical analysis shows that the Weibull parameters, that is, breakdown voltage and shape parameter vary from a distribution to another one. The values of shape parameter are in good agreement with those reported elsewhere.[14,19]

TABLE I
Values of Scale Parameter, Shape Parameter, and Equation of Weibull Plot

	Nominal breakdown voltage (kV)	Shape parameter	Equation of Weibull graph
Before aging	$5.157 < V_b = 5.343 < 5.528$	$5.860 < \beta = 7.320 < 8.642$	$Y = 7.320X - 5.328$
6000 h at 200°C	$1.520 < V_b = 1.754 < 2.009$	$1.451 < \beta = 1.812 < 2.139$	$Y = 1.812X - 0.442$
2700 h at 220°C	$2.504 < V_b = 2.783 < 3.076$	$1.962 < \beta = 2.451 < 2.894$	$Y = 2.451X - 1.090$
1300 h at 240°C	$2.308 < V_b = 2.593 < 2.895$	$1.783 < \beta = 2.227 < 2.629$	$Y = 2.227X - 0.921$
520 h at 260°C	$2.285 < V_b = 2.570 < 2.873$	$1.767 < \beta = 2.207 < 2.606$	$Y = 2.207X - 0.905$
140 h at 280°C	$1.363 < V_b = 1.588 < 1.835$	$1.360 < \beta = 1.699 < 2.006$	$Y = 1.699X - 0.341$

Variation of breakdown voltage versus the aging time

The variation of the nominal breakdown voltage (V_b) versus the aging time is illustrated in Figure 2. The evolution can be described as follows:

- At 200°C, V_b decreases slowly from 5.34 kV to 1.60 kV after an aging time of 6270 h.
- At 220°C, first, V_b increases from 5.34 kV up to 6.91 kV after 1125 h. Then, V_b decreases to 2.38 kV corresponding to an aging time of 2800 h.
- At 240°C, initially, V_b increases from 5.34 kV up to 6.12 kV after an aging time of 460 h. Then, it decreases to 2.03 kV corresponding to 1420 h.
- At 260°C, V_b increases from 5.34 kV up to 5.69 kV corresponding to an aging time of 56 h. Beyond this period, it decreases rapidly until 2.17 kV after 537 h.
- At 280°C, V_b decreases abruptly from 5.34 kV to 1.44 kV after an aging time of 145 h.

As expected, the higher the aging temperature, the shorter the degradation time is. These results are in agreement with the Arrhenius's law. It is well known that the thermal degradation of a majority of polymers can be described by a so-called Arrhenius plot,[20] which is in reality a combination of the Arrhenius's law and the rate of the first or second-order chemical reaction. It is expressed as:

$$\frac{dR}{dt} = k_0 \exp\left(-\frac{E}{kT}\right) \quad (5)$$

where $\frac{dR}{dt}$ is the derivative of the property R with respect to the time; in this study it is in kilovolt per hour, k_0 the quasi-constant, E (J mol^{-1}) the activation energy of the reaction involved, k the Boltzmann constant ($k = 8.314$ J mol^{-1} K^{-1}), and T (K) the absolute temperature.

The evolution of the breakdown voltage can be explained as follows: aging intensifies the thermal agitation, which leads to a progressive reduction in viscosity, expressing a weakening of the molecular bonds and an increase of the free volume. This phenomenon has, like a consequence, an increase in the mobility of charge carriers, which lower the breakdown voltage, whereas the augmentation of this latter is attributed to a crosslinking of the polymer and a diminution in the mean-free path, which decreases the mobility of charge carriers. For an aging time less than 2560 h, the values of breakdown voltage at 220°C are higher than that obtained at 200°C. The increase reaches 69.8%. It can be explained by the fact that at 200°C, large-size defects were created during the aging, which facilitates the initiation and the development of electrical discharges leading to dielectric breakdown. Although at 220°C, the material is crosslinked. After 2560 h aging, the degradation occurs rapidly at 220°C than at 200°C, which is in agreement with Arrhenius' law.

Dependence of the shape parameter on the aging time

Figure 3 (a–e) shows the dependence of the shape parameter on the aging time at the different temperatures. It can be described as follows:

- At 200°C, β decreases from 6.35 to 1.81 after an aging time of 6000 h [Fig. 3(a)].
- At 220°C, at first β decreases from 7.33 to 6.81 then grows until 7.67 after 1232 h. Beyond this time, it diminishes slowly to 7.51. Then, it

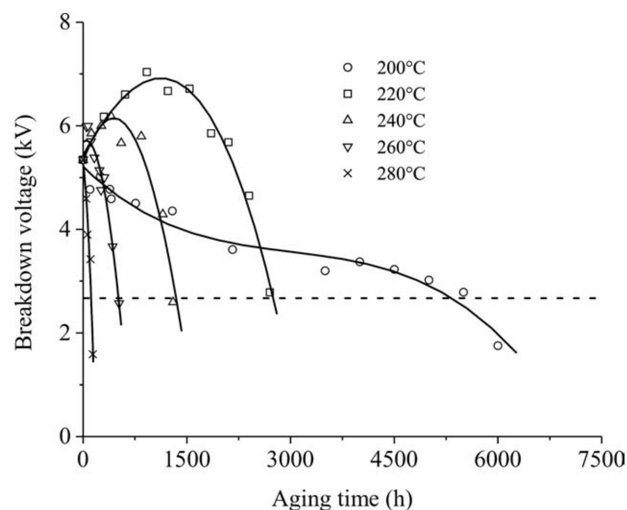

Figure 2 Variation of breakdown voltage in function of aging time at different temperatures. The dotted line corresponds to 50% loss of breakdown voltage.

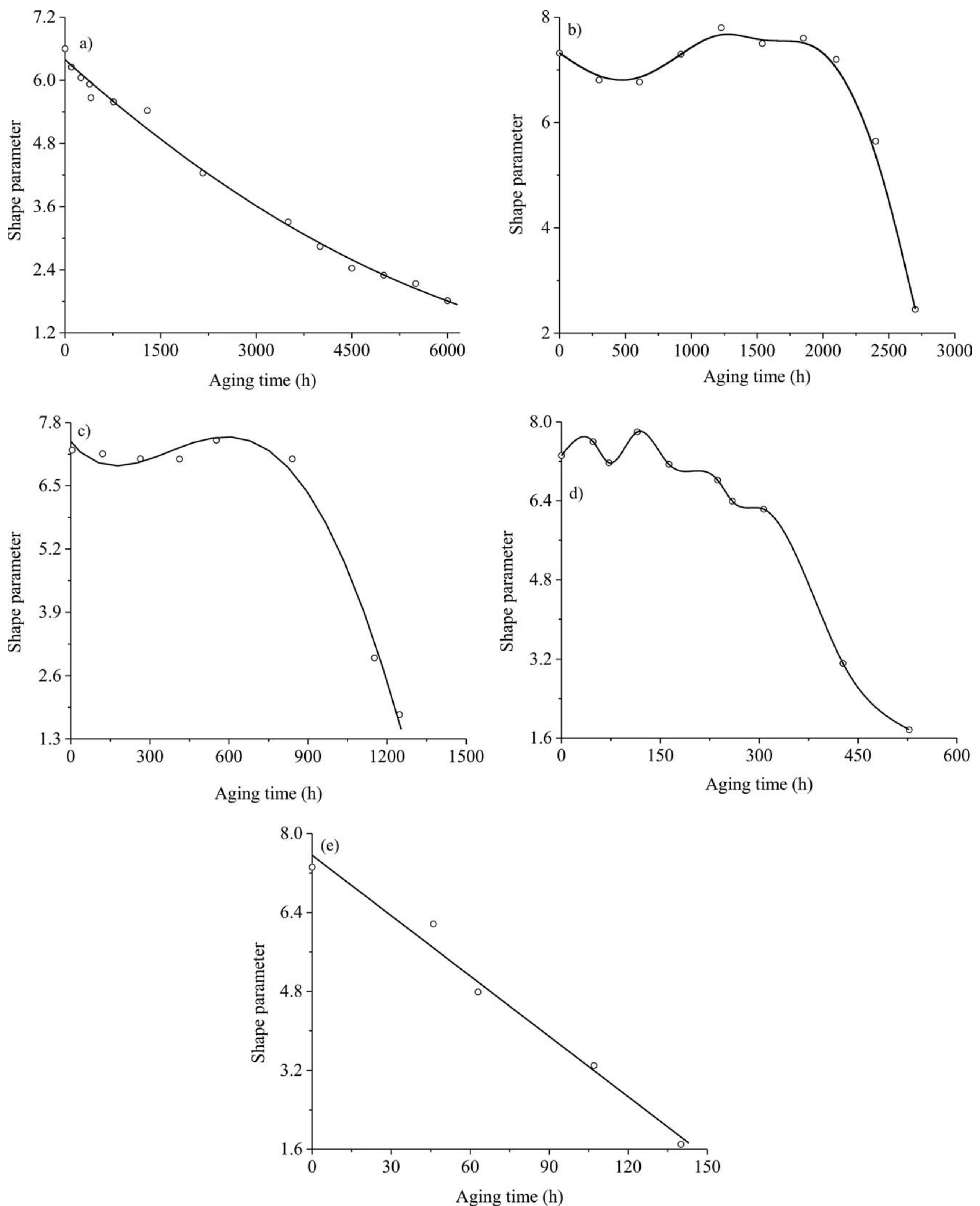

Figure 3 Variation of the shape parameter versus aging time—at 200°C [Fig. 3(a)]; at 220°C [Fig. 3(b); at 240°C [Fig. 3(c)]; at 260°C [Fig. 3(d)]; at 280°C [Fig. 3(e).

decreases abruptly to 2.47 corresponding to 2700 h [Fig. 3(b)].

- At 240°C, at the beginning, β decreases slowly from 7.40 until 6.91 after an aging time of 174 h. Then, it increases up to 7.50 and decreases abruptly until 1.49 after 1256 h [Fig. 3(c)].
- At 260°C, initially β is practically invariable versus aging time. After 180 h, it decreases

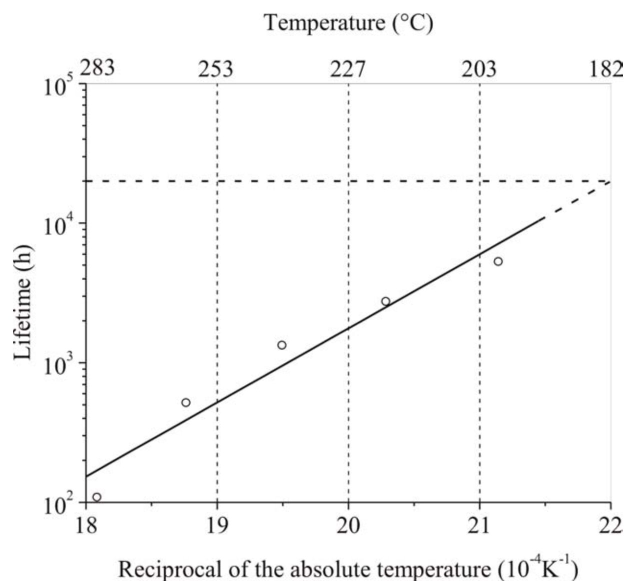

Figure 4 Characteristic of thermal lifetime.

rapidly to 1.77 corresponding to 530 h aging [Fig. 3(d)].

- At 280°C, β decreases rapidly from 7.55 to 1.72 after 143 h [Fig. 3(e)].

The variation of the shape parameter is attributed to the size distribution of the defects present in the dielectric or created during the aging. The decrease of β can be explained by the fact that the defect size increases, whereas the augmentation of β is allotted to the arrangements in the molecular structure of the material. This phenomenon has been reported by Coppard et al.[21] Also, Katsuta et al.[22] demonstrate that the breakdown voltage of XLPE and the shape parameter of the Weibull graphs depend on the kind and the size of defects. These authors considered various defects: voids, contaminants, and protrusions.

Thermal lifetime curve

The lifetime of an insulation system subjected to thermal aging may be written as[20]:

$$L = A \exp\left(\frac{B}{T}\right) \tag{6}$$

with L (h) the lifetime, A the pre-exponential constant, B (K) the constant equal to the ratio E/k, where k is the Boltzmann constant ($k = 8.314$ J/mol/K), E (J mol^{-1}) the activation energy of the chemical reaction controlling the aging mechanism, and T (K) the absolute temperature.

To determine the thermal endurance graph, the degradation criterion was chosen for a 50% loss of the property. Then, the lifetime corresponding to a 50% loss of the breakdown voltage was derived for the different temperatures from Figure 2. The variation of breakdown voltage (63.2%) versus the reciprocal of the absolute temperature is represented in Figure 4. As can be seen, the characteristic is a straight line. The linearity of the thermal endurance graph indicates that the degradation is governed by a first-order chemical reaction. According to the expression (6) the thermal lifetime was derived. It is given by the following expression:

$$t = 4.3x10^{-8} \exp\frac{12222}{T} \tag{7}$$

The corresponding activation energy E is 24.31 kcal/mol. By extrapolating the thermal endurance curve, presented in Figure 4, to a lifetime of 20,000 h, the temperature index was determined. Its value is equal to 182°C.

The chemical reaction rate was calculated using the following relationship:

$$r = \frac{1}{\mathrm{V_b}}\frac{\mathrm{dV_b}}{\mathrm{dt}} \quad (\mathrm{h}^{-1}) \tag{8}$$

with dV_b (kV) the property variation, V_b (kV) the property value, dt (h) the time interval between the beginning of aging, and the degradation corresponding to 50% loss of breakdown voltage.

In this case, the quotient dV_b/V_b is equal to 50%.

The results of the values of chemical reaction speed and the degradation times are shown in Table II.

Mechanism of degradation

According to the results reported by Araki et al.,[3] the thermal degradation of polyesterimide occurs as follows: copper dissolves within the insulation layer and accelerates the degradation of the dielectric. The decomposition of the insulating material happens at the polyesterimide–copper interface. The oxygen can easily diffuse into the polymer layer and attack copper, resulting in the formation of copper oxide. The degradation of polyesterimide is caused by scission reaction of imide bond and ester bond.

Numerous researchers have highlighted that copper accelerates thermal degradation of polymeric

TABLE II
Values of the Chemical Reaction Speed Corresponding to Each Temperature

Aging temperature (°C)	Degradation time (h)	Chemical reaction speed (10^{-4} h)
200	5326	0.9
220	2752	1.8
240	1352	3.7
260	514	9.7
280	117	42.7

materials.[23–25] Ou et al.[23] studied the thermal-oxidative degradation of chitosan-cupric ion compounds in air. The mixing of cupric ion with chitosan has an effect on the formation of new phases of the polymer. Cupric ion leads to the decrease of the crystallinity of chitosan and its thermal instability. Activation energy decreases slightly with increasing weight fraction of cupric ion. Ŝelih et al.[24] mentioned that the addition of copper in cellulose leads to the oxidative degradation resulting to the production of hydroxyl radicals, which has a correlation with chain scission. The activity of copper is catalytic. Sipinen et al.[25] found that the copper is useful in catalyzing the oxidative degradation of polypropylene films at 20–70°C.

In the present investigation, a change in color of the specimens from orange to black accompanied by cracks was observed after aging for all the temperatures. The changing color occurs practically at the middle of aging for the temperatures of 240, 260, and 280°C. For 200 and 220°C, the changing was observed at the end of aging.

CONCLUSION

The obtained results demonstrate that breakdown voltage of polyesterimide is affected by thermal aging. The degradation is accelerated when the temperature is increased. The raise of the breakdown voltage is attributed to a crosslinking that decreases the mean-free path leading to a diminution in the mobility of charge carriers, whereas its decrease is due to the fact that thermal aging decreases the viscosity, expressing a weakening of the molecular bonds and an increase in the free volume, which induces an augmentation of charge carrier mobility. The shape parameter changes with aging time. Its increase is attributed to the arrangements of the molecular structure, whereas its decrease is due to the augmentation in the size of defects. The thermal endurance graph is a straight line, which indicates that the degradation is governed by a first-order chemical reaction. The activation energy and the temperature index are 24.3 kcal/mol and 182°C, respectively. The degradation is governed by the dissolution of copper within the insulating material and accelerates its decomposition occurring at the polyesterimide–copper interface. The oxygen can easily diffuse into the polymer layer and attack copper, resulting in the formation of copper oxide. The decomposition of polyesterimide, caused by scission reaction of imide-bond and ester-bond, is accompanied by a change in color and a presence of cracks.

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