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RAPID COMMUNICATION

Pressure effect on the electrical ageing of polyethylene

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Abstract. The purpose of this study is to show the effect of hydrostatic pressure on the electrical ageing, by a dc voltage, on polyethylene used in submarine cables. Experimental results are obtained at 1 and 300 bar, for a temperature of 70 °C in the range of an electrical field of 1.1–4.5 MV cm⁻¹ and for up to 1500 h of ageing. By using the Weibull statistic and the estimation of confidence bounds of 90%, the results have shown an increase of lifetime with pressure. The time dependence of the breakdown behaviour of the two materials (HDPE and XLPE) is consistent with the inverse power law model. The pressure effect may be explained by changes in the activation free energy for the formation of submicrocavities.

1. Introduction

In our previous work on the electrical behaviour of insulating material such as cross-linked polyethylene (XLPE) and high-density polyethylene (HDPE), we have shown that for short time tests, with a rise time of 4 kV s⁻¹, the breakdown field decreases with temperature from 20 to 90 °C and increases with pressure in the range of 1–500 bar [1, 2]. To obtain the lifetime behaviour of these polymers in the insulation of submarine power cables under a dc voltage, long time tests, up to 1500 h, were executed for the temperature of 70 °C and the pressures of 1 and 300 bar. These can be considered as the real use conditions of these cables. In this case, the hydrostatic pressure is simulated to the depth of immersed cables.

2. Experimental procedure

The samples were 100 μm thick films of HDPE and XLPE. The samples were placed into the measuring cell composed of a system of high-voltage relays and ten pairs of electrodes made with stainless-steel, which followed a Rogowsky profile.

At atmospheric pressure ($p = 1$ bar), the measuring cell was immersed into a chamber filled with silicone oil to avoid flashover and partial discharge effects.

For high-pressure measurements, $p = 300$ bar, the experimental chamber was a high-pressure bomb with a working volume of 9500 cm³ [3]. The maximum pressure of 1500 bar is given by a high-pressure compressor using gaseous nitrogen as a transmitting fluid. The desired temperature could be obtained by a heating resistance system of 1600 W.

The dc voltage, ranging from 15 to 50 kV, was provided by a generator of 100 kV and was applied to a batch of 10 samples, with a maximum duration of 1500 h. During this period, when a dielectric breakdown occurred in one of the samples, a positive pulse was generated across the detection system, the applied voltage was immediately removed for this sample and the breakdown time value t_i was recorded while the other samples were continuously submitted to the applied voltage.

3. Results and discussion

All of the results obtained were analysed with a Weibull distribution, or more specifically, the two parameter Weibull distribution [4].

The well known expression of the two parameter Weibull distribution is given by

$$F(t) = 1 - \exp(-(t/t_0)^\beta)$$

where $F(t)$ is the probability of failure, t_0 is the scale parameter ($t_0 > 0$) and represents the time for $F(t)$ to reach 63.2%, β is a shape parameter and a measure of the spread of the data and t is a random variable (the time in our case).

To evaluate the data using the Weibull distribution a special procedure has to be followed:

- (i) The data (breakdown times) must be ordered from smallest to largest and the probability P_i (in per cent) calculated for each time t_i using the relationship

$$P_i = F(t_i) = 100\{0.69/n + (i - 1)[1/(n - 1) - 1.38/(n(n - 1))]\}.$$

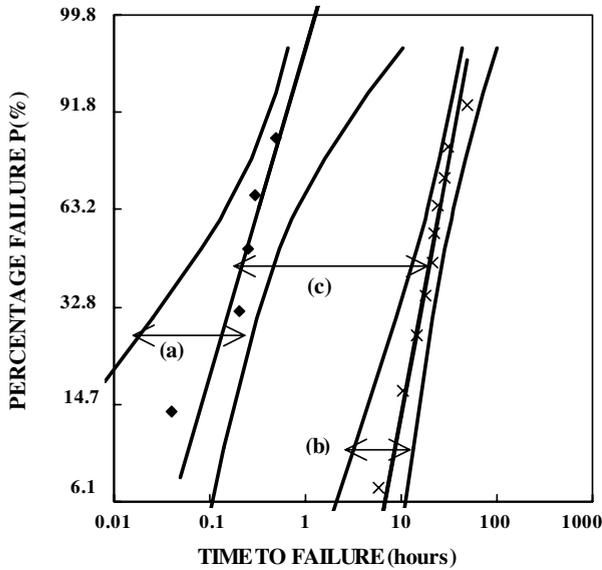


Figure 1. Weibull plot of the ageing data of HDPE at 2.5 MV cm^{-1} and 70°C : (a) confidence bounds at 90% level (data at 1 bar), (b) confidence bounds at 90% level (data at 300 bar) and (c) best-fit lines of the Weibull distribution. In the figure \blacklozenge denotes a pressure of 1 bar and \times denotes a pressure of 300 bar.

- (ii) The experimental points $(t_i, \ln 1/(1 - P_i))$ are plotted on logarithmic scale.
- (iii) On the graph from (ii) the best-fit line of the Weibull distribution is plotted. This line is determined by the estimation of the values of t_0 and β , based on the maximum-likelihood method.
- (iv) The last stage is to determine, using this graph, the limits of the confidence intervals at the different percentiles.

The use of the Weibull distribution shows that the confidence intervals for the level of confidence value of 90% do not overlap at different percentiles, shown in figures 1 and 2, in which the two lifetime distributions, at 1 bar and 300 bar of our insulators, are significantly different.

To explain the effect of pressure on the ageing of the material we try to apply the two following theoretical models: the inverse power law model [5] and Crine's model [6].

3.1. Compatibility with the inverse power law model

For the inverse power law model, all the values of the breakdown field E and time t obtained with $p = 1$ and 300 bar and $T = 70^\circ\text{C}$ for the HDPE and the XLPE samples can be represented on the logarithmic scale ($\log E, \log t$). Figures 3 and 4 show that these results correspond to the inverse power law model that follows the empirical expression:

$$E^n t = k$$

where k is a constant and the exponent n is the voltage endurance coefficient whose value depends on the material quality.

From these curves we can conclude that the dielectric behaviour of the HDPE samples is better than that of the XLPE samples. For example, to obtain a breakdown time $t = 100 \text{ h}$ at $p = 300 \text{ bar}$, $E(\text{HDPE}) = 2.4 \text{ MV cm}^{-1}$ and

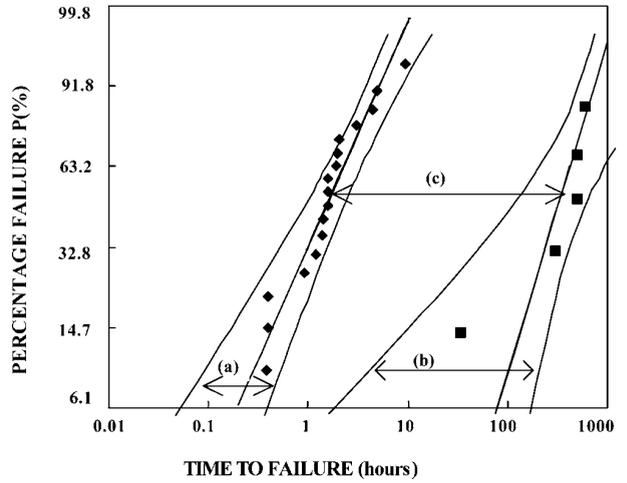


Figure 2. Weibull plot of the ageing data of XLPE at 1.5 MV cm^{-1} and 70°C : (a) confidence bounds at 90% level (data at 1 bar), (b) confidence bounds at 90% level (data at 300 bar) and (c) best-fit lines of the Weibull distribution. In the figure \blacklozenge denotes a pressure of 1 bar and \blacksquare denotes a pressure of 300 bar.

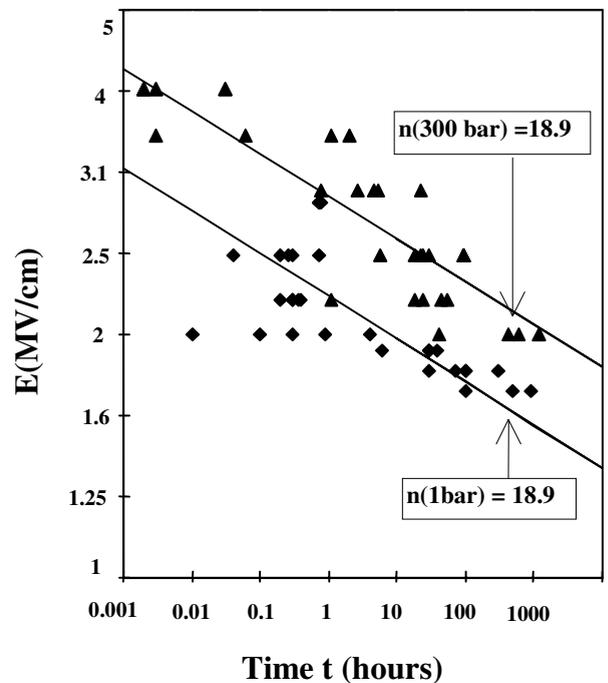


Figure 3. Representation of the ageing data of HDPE on a logarithmic scale for a sample aged at 70°C , according to the inverse power law model. In the figure \blacklozenge denotes a pressure of 1 bar and \blacktriangle denotes a pressure of 300 bar.

$E(\text{XLPE}) = 1.7 \text{ MV cm}^{-1}$. This can be shown by the values of n ; for example, for $p = 300 \text{ bar}$ $n(\text{HDPE}) = 18.9$ and $n(\text{XLPE}) = 15.4$.

Although commonly used, the inverse power law model can only give information on the variations of the lifetime with such or such parameter (pressure, temperature, etc), but not on the origin of the breakdown processes.

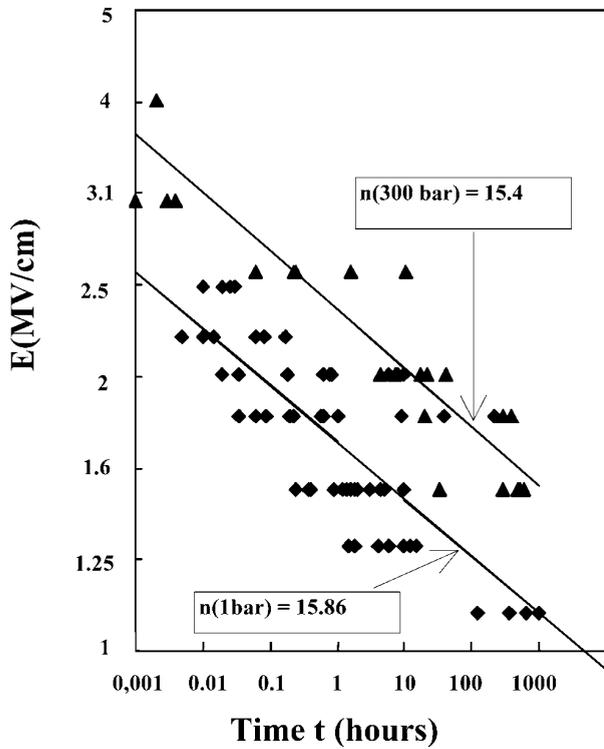


Figure 4. Representation of the ageing data of XLPE on a logarithmic scale, for a sample aged at 70 °C, according to the inverse power law model. In the figure \blacklozenge denotes a pressure of 1 bar and \blacktriangle denotes a pressure of 300 bar.

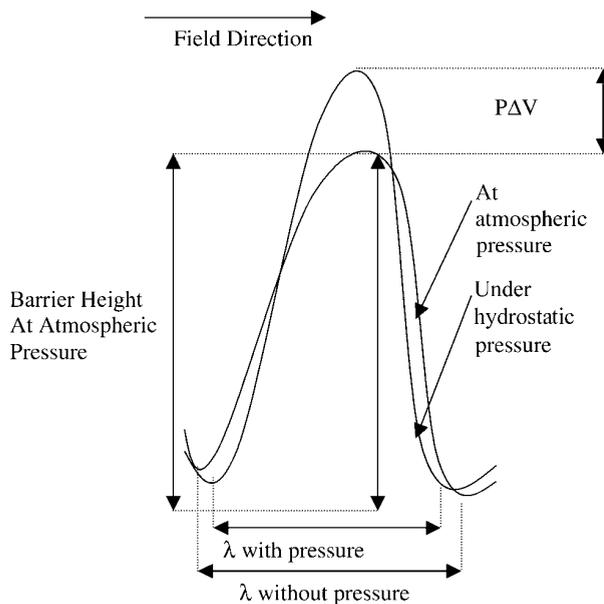


Figure 5. Variations of the barrier height ΔG and the barrier width λ with pressure.

3.2. Application of Crine's model

Among the different ageing models, we attempted to apply Crine's model [6] to explain our results. The model proposed that the electrically-induced stresses may result in local overstressing and therefore in the localized heating of the polymer chains. This will ultimately result in the

Table 1. Calculated values of ΔG , λ and E_c according to Crine's model for the ageing of HDPE and XLPE at 70 °C.

Material	Pressure (bar)	ΔG (eV)	λ (Å)	E_c (MV cm ⁻¹)
HDPE	1	1.58	19.5	0.33
	300	1.61	16.1	0.44
XLPE	1	1.39	16.5	0.38
	300	1.51	15.6	0.45

rupture of the overstressed segments which would initiate a submicrocavity. This is an irreversible ageing process.

In this model, the energy parameter controlling the rate of ageing is the activation free energy ΔG , which is equal to $\Delta H - T\Delta S$, where ΔH and ΔS are the activation enthalpy and entropy, respectively. Under the action of electric field E the height of the energy barrier is reduced by a factor $\text{csch}(e\lambda E/KT)$ where λ is the width of the barrier. The lifetime t is described by [7, 8]

$$t = h/KT \exp(\Delta G/KT) \text{csch}(e\lambda E/KT)$$

where h and K are the Planck and Boltzmann constants, respectively.

In the high-field region, above a critical field E_c , this equation reduces to the exponential dependence:

$$t = h/KT \exp(\Delta G - e\lambda E)/KT.$$

With this equation, the values for activation free energy ΔG and the width of the barrier λ can be obtained from the plot of $\log t$ against E , and may also be used for the lower-field region. Table 1 gives the results of application of this model to our studied insulators.

The increase of ΔG is due to that of ΔH which depends on the pressure p from the term $p\Delta V$ as follows: $\Delta G = \Delta H - T\Delta S = \Delta E - T\Delta S + p\Delta V$ ($p\Delta V$ is generally negligible comparative to ΔE at atmospheric pressure [9]).

For the decrease of λ with p , we assume that the pressure effect is to compress the amorphous phase of the polymer, thereby reducing λ (λ could be considered as the average length of the amorphous phase of polymer [7, 8]).

These hypotheses are shown in figure 5.

4. Conclusions

The experimental measurements carried out at pressures of 1 and 300 bar, at a temperature of 70 °C, on HDPE and XLPE samples show that the pressure delays the electrical ageing. The results were analysed by the Weibull's statistic with a confidence bounds at a 90% level.

The well-known empirical inverse power law model gives a qualitative analysis. For example, for the XLPE sample with a field of 1.5 MV cm⁻¹, the lifetime $t(1 \text{ bar}) = 2 \text{ h}$ and for $t(300 \text{ bar}) = 200 \text{ h}$, for the HDPE sample at an electrical field of 2.5 MV cm⁻¹, the lifetime $t(1 \text{ bar}) = 0.2 \text{ h}$ and $t(300 \text{ bar}) = 20 \text{ h}$.

The breakdown process could be explained by the Crine's model based on the existence of submicrocavities in the material with the dependence of the activation free energy ΔG and the barrier width λ on pressure effects.

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