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QUANTIFICATION OF KRAFT PAPER AGEING IN MINERAL OIL IMPREGNATED INSULATION SYSTEMS THROUGH MECHANICAL CHARACTERIZATION

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ABSTRACT

Most power transformers use Kraft paper as the main solid insulation between the winding conductors. Dielectric oil used in transformers as an insulating and cooling fluid typically has an operating temperature range of 60-90°C. These service temperatures can cause slow degradation of both the oil and the insulating paper winding, with a loss of mechanical and dielectric properties. In this sense, this paper proposes the possibility of analysing paper degradation through the loss of its mechanical properties. An accelerated thermal ageing of the paper in mineral oil was carried out at temperatures of 110, 130 and 150°C over different periods of time, in order to obtain information on the kinetics of the ageing degradation of the paper. The evolution of the mechanical properties and micro mechanisms of paper failure are analysed as a function of temperature and ageing time. Finally, the results obtained are compared with the traditional method of degradation analysis, based on the degree of polymerisation (DP) measurement.

KEYWORDS: Kraft paper, power transformer, thermal ageing, degradation, tensile test, degree of polymerisation

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INTRODUCTION

Electric power systems are of great importance in the daily life of modern societies and their reliability during operation depends on the lifespan of their components (Zhuravleva et al. 2016). Power transformers are one of these critical elements since they carry out essential functions in the generation and transmission of power (Perrier et al. 2016). Most of the transformers used in electric power systems are currently oil immersed transformers that also contain a solid insulation. The oil used in this type of transformers not only provides a cooling function, but also an electric insulating function together with the dielectric solid materials. This type of transformers can contain up to 12 tonnes of paper insulation wrapped around the windings and 40 tonnes of insulating oil. Their operation temperature is usually between 60-90°C and can have a lifespan of up to 40 years.

The most widely used dielectric liquid in power transformers is mineral oil, obtained from crude oil which has shown suitable thermal and dielectric properties to perform their cooling and insulation functions respectively (Fofana 2013). At the same time, Kraft paper (90% cellulose, 6-7% lignin and 3-4% pentosans) (Emsley and Stevens 1994) is by far the most widely used solid material due to economic factors and ease of manufacture, although other materials are being used to insulate the areas where the operating temperature is believed to be high (Ding and Wang 2008).

Cellulose insulating materials have desirable physical and chemical properties for use as electrical insulators, although they degrade over time. Therefore, the degradation of cellulose materials is a critical factor that determines the life expectancy of a transformer (Prevost et al. 2006). During transformer operation, the dielectric solid undergoes a slow decomposition with loss of its mechanical strength due to the effects of temperature, electrical and mechanical stresses, and the chemical reactions that occur over time. It is commonly established that the main factor influencing the degradation of paper insulation is the temperature (Oommen and Prevost 2006; Prevost et al. 2006). During the operation of the transformer, not only does the dielectric paper undergo continuous degradation, but also the oil progressively loses its dielectric and thermal properties, affecting the insulating and cooling systems of the machines. For this reason, it is essential

to characterise the degradation of the paper and the oil. However, although the state of the oil in a power transformer can be evaluated directly because sample-taking is easily accessible, the sampling of a piece of paper from the windings of transformers in service is not possible.

Thus, non-destructive techniques are used to determine the paper's state. These techniques are based on the measurement of by-products derived from the deterioration of cellulose (furanic compounds, water, CO₂, CO, etc.) which can be used to indirectly evaluate the condition of the insulation paper, once they have dissolved in the oil. Nevertheless, these techniques, which are indirect methods to determine cellulose degradation, have the disadvantage that they depend on the knowledge of the history of the transformer and its components (Emsley and Stevens 1994; Hill et al. 1995; Levchik et al. 1997; Heywood et al. 2000; Kalariya et al. 2007).

Since the lifetime of a transformer depends fundamentally on the state of the solid insulation, as has been shown by different studies (Emsley and Stevens 1994; Heywood et al. 2000; Carcedo et al. 2016), a detailed understanding of the mechanisms of cellulose degradation is needed to predict the remaining life of the insulating paper and give an early warning of premature failure, (Hill et al. 1995). One way to define the kinetic models of solid insulation degradation is through the post-mortem analysis of power transformers, since this type of study attempts to relate the results obtained by the indirect methods of diagnosis described above with the actual state of the components of the machine (Knoch et al. 2007; Prevost et al. 2007; Martins et al. 2011; Leibfried et al. 2011).

However, these life predictions from actual data collected from the transformers in service are complex because there is a large number of operational variables involved during transformer performance and additionally, they are extremely disperse (Madavan and Balaraman 2016). In view of this, laboratory tests are used to provide a strict control of the experimental variables.

Furthermore, these laboratory experiments, performed at constant temperature, need several months to produce results because they are carried out at higher temperatures than service ones. These laboratory tests are used to provide an overview of the chemical process associated with the ageing of the paper (Hill et al. 1995 part1; Hill et al. 1995 part2; Hill et al. 1996; Levchik et al. 1997; Gasser

et al. 1999; Murugan and Ramasamy 2015; Carcedo et al. 2016) and they considerably reduce the economic costs.

The method most commonly used in the literature for characterising the degradation of the dielectric paper involves the determination of the degree of polymerisation (DP) through the measurement of the viscosity. The bond breaking of the major cellulose chains caused by the deterioration results in a decrease in the average molecular weight of the chains. This fact not only causes variations in the physical properties of the oil (generating a significant amount of water, furans, CO and CO₂) (Hill et al. 1995 part1; Hill et al. 1995 part2; Hill et al. 1996; Levchik et al. 1997; Gasser et al. 1999), but also it has been shown that paper undergoes a decrease in its mechanical properties (Lundgaard et al. 2007). For this reason, several studies have proposed models that establish the relationship between the variation of the tensile strength and the degree of polymerisation (DP) over time (Hill et al. 1995 part2, Hill et al. 1996, Gasser et al. 1999).

Other more recent studies (Mirazie et al. 2009; Hoom et al. 2010; Abelmalik et al. 2013; Rodriguez-Celis et al. 2015; Widyanugraha et al. 2015) have performed the comparison between the variation of the tensile strength and different parameters associated with the properties of the oil. For example, one of these parameters is the content of gases dissolved in the oil as: CO, CO₂, H₂, CH₄, C₂H₄, C₂H₆ and C₂H₂. Studies like the one carried out by Hoom (Hoom et al. 2010] have obtained mathematical models that relate the tensile strength with the content of CO and furans dissolved in the oil as a function of the ageing temperature.

Other studies (Mirazie et al. 2009; Abelmalik et al. 2013) have related the evolution of the breakdown voltage in oil and paper, acidity and water content in the oil with the rupture stress, making a comparison between the results obtained from both parameters over time and at different ageing temperatures. There are also works such as the one carried out by Ding (Ding and Wang 2008) in which mathematical models have been proposed for the calculation of the degradation experienced by the insulation paper as a function of time and temperature through the measurement of the DP and tensile index.

However, in all these studies, only strength or tensile index have been analysed, not taking into account the evolution of other important mechanical properties such as strain or energy consumed per unit volume, which are closely related to the brittleness of the material.

Considering the significance that other mechanical parameters might have in the analysis of paper degradation, this paper attempts to correlate the variations suffered by different mechanical properties (strength, Young's Modulus, yield stress, energy consumed, etc.) with the degree of degradation of Kraft paper. The final aim is to establish whether data obtained from the stress-strain curve might be able to provide a new method which can predict dielectric paper failure. Additionally, a mathematical model has been defined based on the degree of polymerisation to predict the remaining life of Kraft paper. The information provided by this method has been compared with that based on the stress-strain curve. This will be used to observe which of the two methods supplies the more useful information and whether the end-of-life criteria of the dielectric material established until now are the most suitable for guaranteeing the reliability of the solid insulation.

MATERIAL

This work has studied the behaviour of Kraft paper, whose properties are gathered in Table 1. Kraft paper was cut into strips of 250 mm in length and 15 mm wide. Due to paper anisotropy, these strips were cut with different fibre direction angles (longitudinal, transverse and 45°). This work aims to assess the degradation rate of Kraft paper immersed in mineral oil (Table 2) taking into account paper anisotropy through mechanical analysis.

Table 1. Kraft paper properties.

Property	Units	Value
Grammage	g/m ²	149.3
Thickness / 5 sheets	µm	198
Apparent density	kg/m ³	754
Moisture	%	6.3
Tensile Index	Nm/g	108.4
Ash	%	<0.6
Aqueous extract conductivity	mS/m	1.5

Dry breakdown strength in air	kV/mm	8.9
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Table 2. Mineral oil properties.

Property	Units	Value
Viscosity, 40°C	mm ² /s	7.6
Viscosity, -30°C	mm ² /s	730
Pour point	°C	-63
Flash point	°C	154
Water content	mg/kg	<20
Breakdown voltage	kV	40-60
Acidity	mg KOH/g	<0.01
Density, 20°C	kg/dm ³	0.877

EXPERIMENTAL METHODOLOGY

Thermal ageing

Kraft specimens were placed into a stainless steel vessel which has a volume of 1 litre and two connections, one for the vacuum pump and another for the vacuumometer, as shown in Fig. 1.



Fig. 1: Vessel for thermal ageing

Once the vessel was closed, it was connected to a vacuum pump until reaching approximately 1 mbar. It was then placed in an oven at 100°C for 24 hours, providing samples with a moisture content of 12%.

After that, 750 ml of mineral oil were introduced into the vessel with a nitrogen headspace of 25% by volume. The thermal ageing was then carried out at different temperatures: 110, 130 and 150°C, and different samples of paper were taken periodically to analyse their properties.

Traditional characterisation of cellulose degradation

The method most commonly used for characterising cellulose degradation involves the determination of the chain scission number as a function of the degree of polymerisation (DP). The degree of polymerisation was determined according to ASTM D4243 by measuring the kinematic viscosity of the paper in solution (the viscosity is related to the molecular weight by the Mark-Houwink Sakurada equation). The viscosity-average DP was obtained based on measurements at 20°C using an automatic viscometer equipped with a two-sphere Ubbelohde tube. Each paper strip was first de-oiled using distilled hexane. Subsequently, the paper was dissolved in a solution of deionised water and bis(ethylenediamine) copper (II) hydroxide which is the viscosimetric solvent. After dissolution of the paper in the prepared solution, its specific viscosity was determined. From this result the intrinsic viscosity of the solution was deduced, and from this data the DP was obtained. For this, it is necessary to know the moisture of the sample, which was determined using Karl Fischer titration.

Tensile characterisation of Kraft paper

For tensile testing, a universal servo hydraulic test machine was used with an axial load cell of ± 1 kN capacity, an actuator of ± 50 mm of dynamic stroke and equipped with pneumatic flat grips. The ends of the paper strips were protected with adhesive paper to prevent the grips from causing any damage to the paper. The length of the paper strips for the measurement of the strain was set at 180 mm and the rate of separation of the grips was set at 20 mm / min until the specimen rupture, according to ISO 1924-2 2009. The parameters obtained in the test were

Young's Modulus, E , yield stress, σ_y , rupture strength, σ_R , strain under ultimate strength, ϵ_{cm} , and energy consumed per unit volume of the failure zone, ER .

Fractographic analysis

Finally, the fractured surfaces of the tensile tested paper samples were analysed by a scanning electron microscope (SEM), brand Carl Zeiss, model EVO MA15, to identify the failure mode after different stages of thermal ageing.

ANALYSIS AND RESULTS

Any attempt to characterise Kraft degradation should satisfy two requirements. Firstly, it has to include the essential physical aspects of the degradation process. Secondly, its mathematical expression must be in agreement with the experimental data. This work has started with the development of a cellulose degradation equation, expressed in terms of DP, and has followed with the study of mechanical characterisation. This could be an alternative method which offers a more detailed analysis of dielectric paper degradation.

Paper ageing model

Figure 2 shows the evolution of the degree of polymerisation, DP, as a function of the ageing time and the temperature at which thermal ageing took place. It can be seen that the DP decreases over time and this decrease becomes faster when the ageing temperature increases.

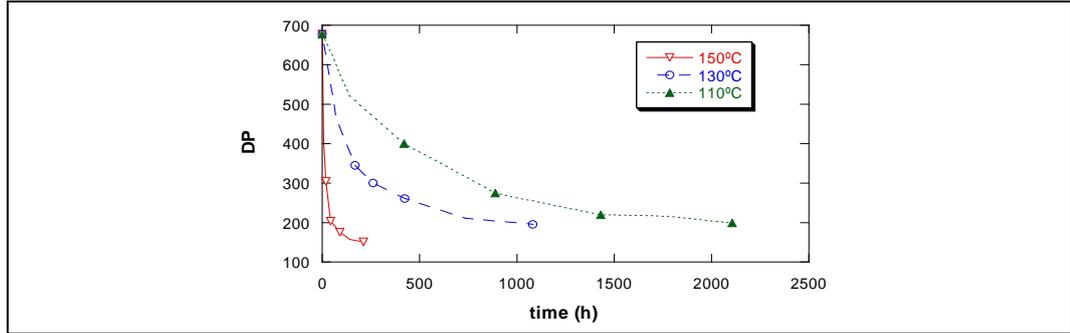


Fig. 2: Evolution of the DP as a function of time and temperature

In order to define a paper deterioration model based on $DP = f(t, T)$, the damage parameter D is defined based on Eq. 1,

$$D = 1 - \frac{DP_i}{DP_0} \quad (1)$$

The damage can be obtained from the DP_i value in any situation of time (t) and temperature (T) and from the DP_0 , which is the value of the DP of the original paper not subject to ageing ($DP_0 = 678$). In this way, the paper without ageing will present no damage, $D = 0$. Figure 3 shows the evolution of the damage parameter, D , with t for different ageing conditions. This figure shows the good correlation between the experimental data and the adjustment performed according to Eq. 2.

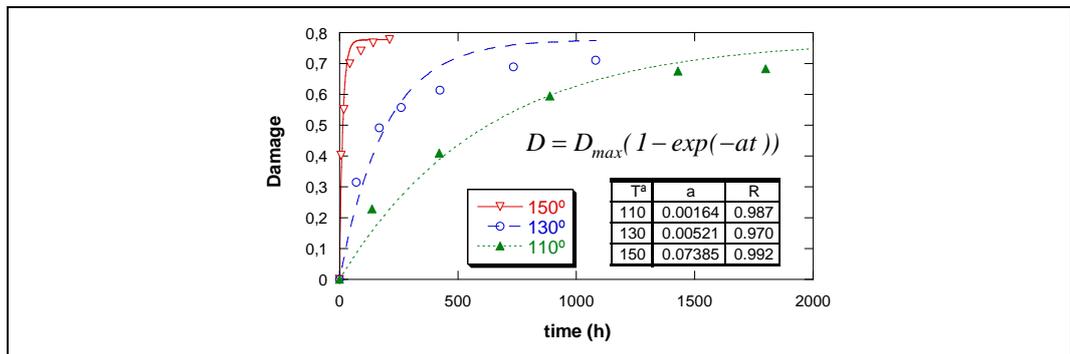


Fig. 3: Evolution of the damage, D , in function of the t and the T

$$D = D_{max}(1 - \exp(-at)) \quad (2)$$

where the constant a depends on the ageing temperature and D_{max} is the maximum value reached experimentally by the damage, $D_{max} = 0.777$. The constant a can be expressed by means of an exponential law Eq. 3 as a function of the ageing temperature.

$$a = 3,626 \cdot 10^{-8} \cdot \exp(0.0952T) \quad (3)$$

Finally, the degree of polymerisation can be expressed by means of Eq. 4 as a function of the time and the temperature.

$$DP_i = DP_0(1 - D_{max}(1 - \exp(-at))) \quad (4)$$

Figure 4 compares the experimental values with those obtained by the behaviour model for the temperatures under study, as well as the theoretical behaviour for other temperatures, including those typical of the operation of power transformers.

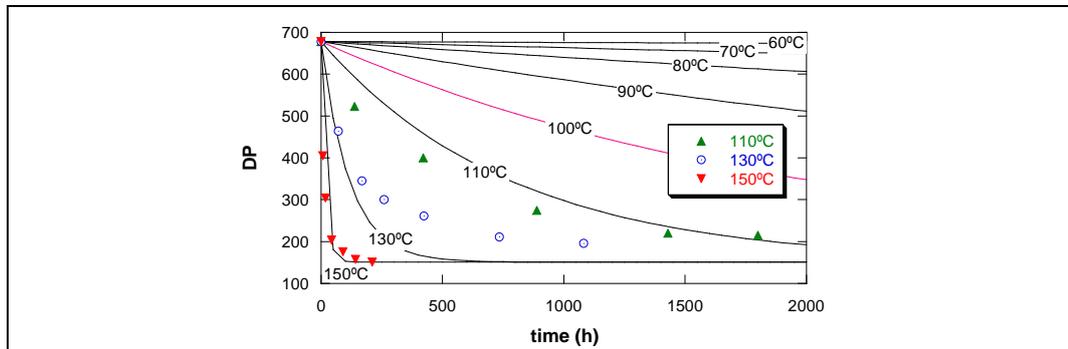


Fig. 4: Comparison of the DP behaviour model and experimental values

A critical degree of polymerisation $DP_c = 200$ is often used as the end-of-life criterion (Lundgaard et al. 2007). Using Eq. 5, if the DP_c is entered as D_i , the estimated life time is obtained as a function of the operating temperature of the transformer, Fig. 5. It can be verified that for a temperature of 60°C , under normal conditions of operation, the estimated life time is about 25 years and as the ageing rate increases, the time decreases logarithmically.

$$t_i = -\frac{1}{a} \cdot \ln\left(1 - \frac{D}{D_{max}}\right) \quad (5)$$

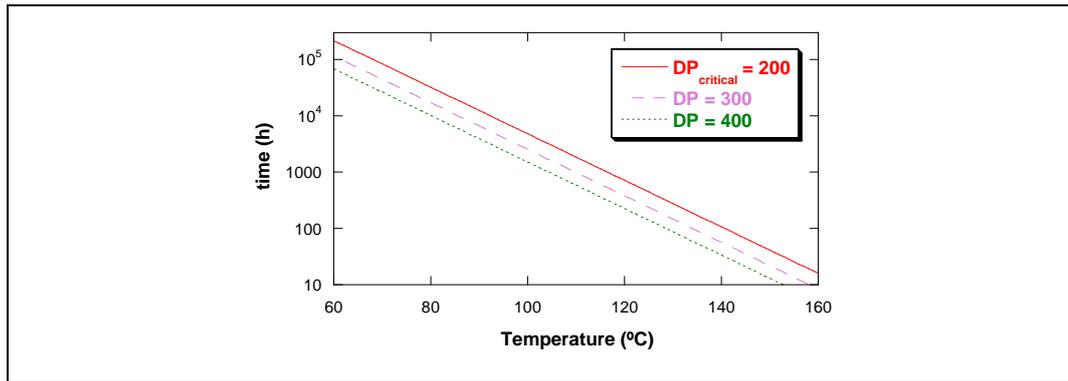


Fig. 5: Estimated time of life as a function of the temperature

Alternative method to evaluate the degradation degree of the paper

When the mechanical behaviour of the original paper is analysed as a function of the fibre direction angle, gathered in Fig. 6, a strong anisotropy can be verified. It can be observed that the rupture strength, when the paper fibres are in the same direction as that with which the test machine applies the load, is two times the strength obtained when the fibres are in cross direction to the test machine, while the strain is half. The strength values when the fibres are at 45° are intermediate and are discarded for the rest of the study.

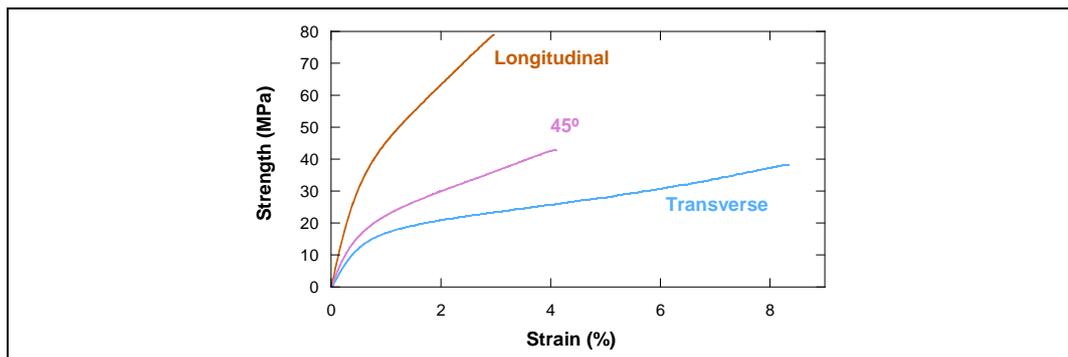


Fig. 6: Anisotropy of Kraft paper

Figure 7 shows the evolution of the strength-strain curves obtained for different ageing times and an ageing temperature of 150°C.

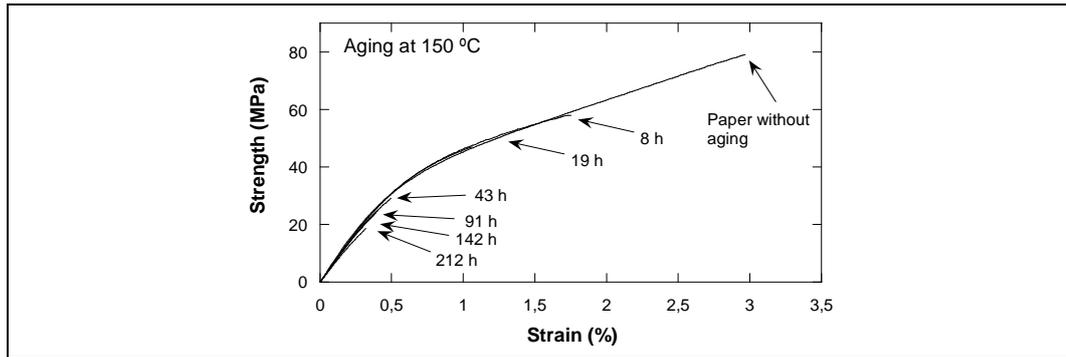


Fig. 7: Influence of the t of ageing on strength

It can be verified in Fig. 7 that the curves are practically coincident and the rupture points have to be indicated on the graph to distinguish them. In view of the results, it can be observed that the Young's Modulus can hardly provide any information on the degree of degradation, since it is practically the same in all the curves and only in those states of severe degradation does it show a slight decrease. As for the yield stress, σ_y , in more than half of the analysed samples its value is the same as that of the rupture strength, due to the fragility of the most deteriorated samples. It will be the parameters of rupture strength and strain under ultimate strength that clearly depend on the paper degradation.

Figure 8 shows the evolution of these two parameters with the time of ageing for the three ageing temperatures analysed. The rupture strength and strain under ultimate strength are considerably affected by time and temperature. Regarding the energy consumed in the rupture, ER , which is a combination of strength and strain, time and temperature have a greater effect, as can be seen in Fig. 9. It can be observed in Figs. 8 and 9 that there is an important linear decrease in the properties at the beginning of thermal ageing and a later stabilisation of the values of the analysed properties, but with relatively poor values. In addition, the loss of properties is observed in a similar way when fibre orientation is longitudinal and transverse.

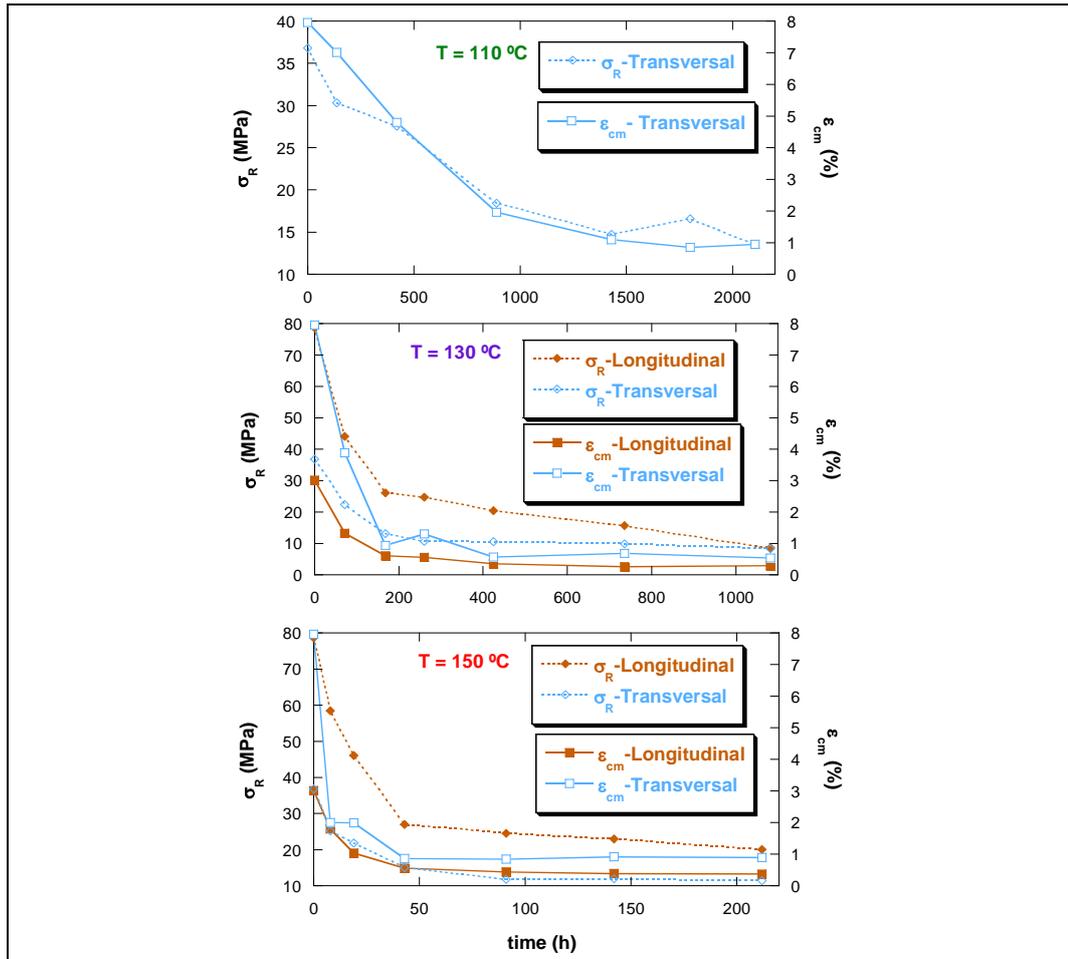


Fig. 8: σ_R and ϵ_{cm} as a function of the ageing t

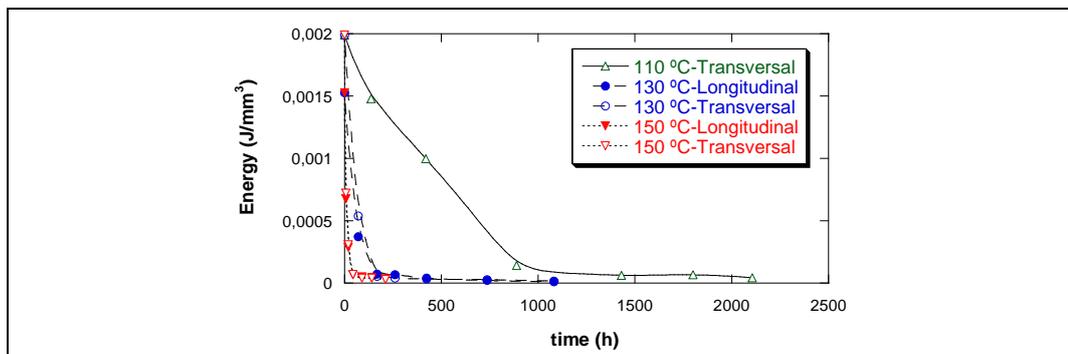


Fig. 9: E_R as a function of ageing time and temperature

It is verified from the two figures above that the critical ageing times, t_c , for which the maximum loss of mechanical properties is produced, are 30, 175 and 1000 hours for the ageing temperatures of 150, 130 and 110°C, respectively. These critical times represent the period required to obtain $DPC = 200$. It can be observed in Fig. 10 that before reaching the DPC, a DP of around 300, the mechanical properties are already minimal. Therefore, it might be of interest to

redefine the critical values of DP (200) and strength (50% of initial value), as it is possible that the general failure mode of the Kraft changes before obtaining these values of DP and σ_R .

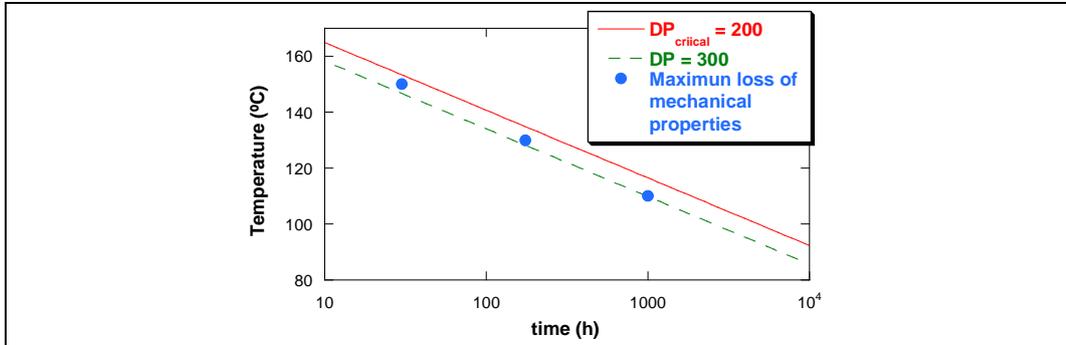
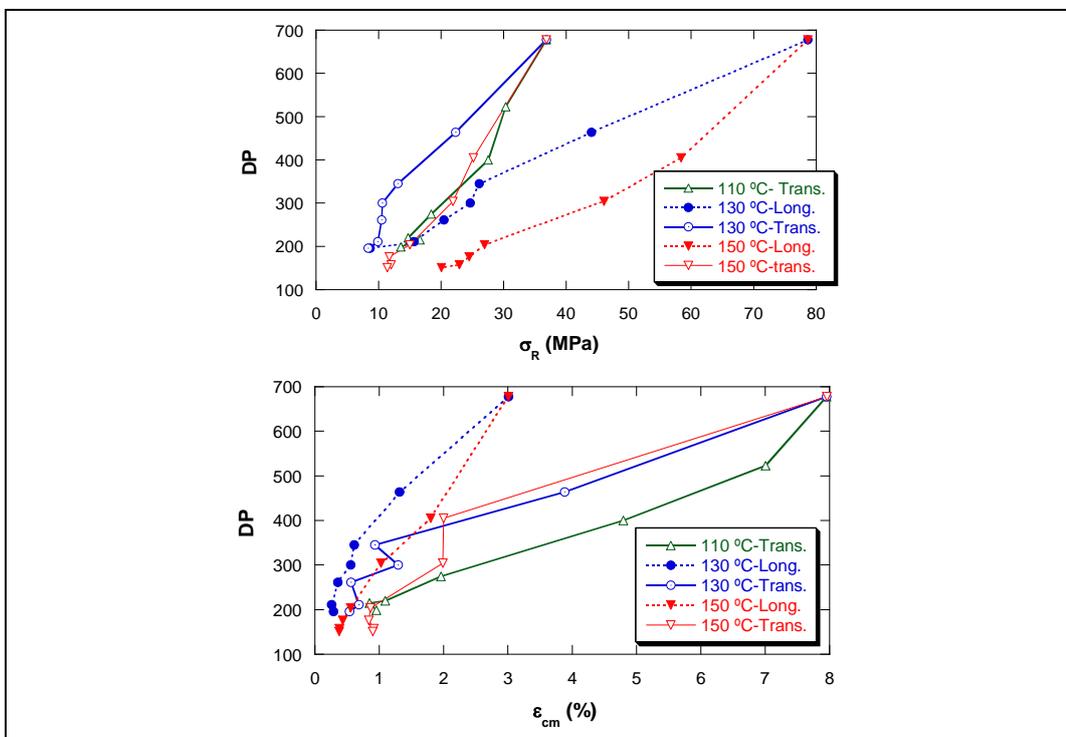


Fig. 10: Comparison between t_c and DP_c

Figure 11 gathers the correlation between the degree of polymerisation and the three variables obtained in the tensile tests, σ_R , ϵ_{cm} and ER. It is possible to verify the existence of a linear relationship between these parameters and DP up to the value close to 300. After this degree of degradation, the mechanical properties hardly change.



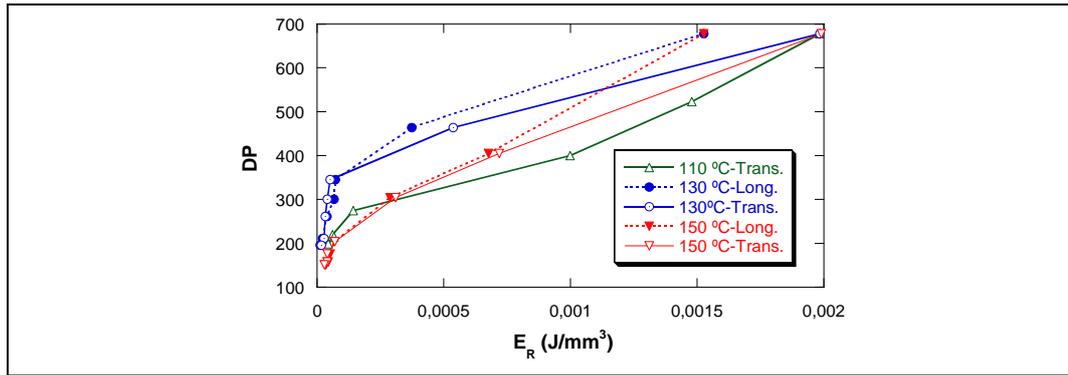


Fig. 11: DP vs test tensile parameters

Fractographic analysis

Fractography can be defined as the study of the fractured surfaces of materials. It is routinely used to determine the cause of failure through the study of the fractured surface characteristics. This technique can be used as a quick and simple procedure to determine the root cause of material failure (Greenhalgh and Hiley 2008). In some cases, fractography requires examination at a finer scale which is usually carried out using a Scanning Electron Microscope (SEM). This device has been used in this work to evaluate the rupture section of Kraft strips.

In the fractography of Fig. 12, it can be verified that the fibres in the rupture section have undergone significant strain when the Kraft paper is new. However, once the paper has been aged this strain is reduced, even at 72 h (Fig. 13). And after a considerable time of ageing, when DP is close to 200, the rupture is totally fragile (Fig. 14), with no strain in the fibres. This coincides with the macroscopic behaviour shown in the tensile tests. It has been verified that the failure mode has changed and this is critical, because when the behaviour of a material is fragile the crack initiation, propagation and final fracture occur instantly and the probability of partial discharges or short circuits would increase in the power transformer.

Therefore, it is essential to define mathematical models which relate the macroscopic properties with the failure mode.

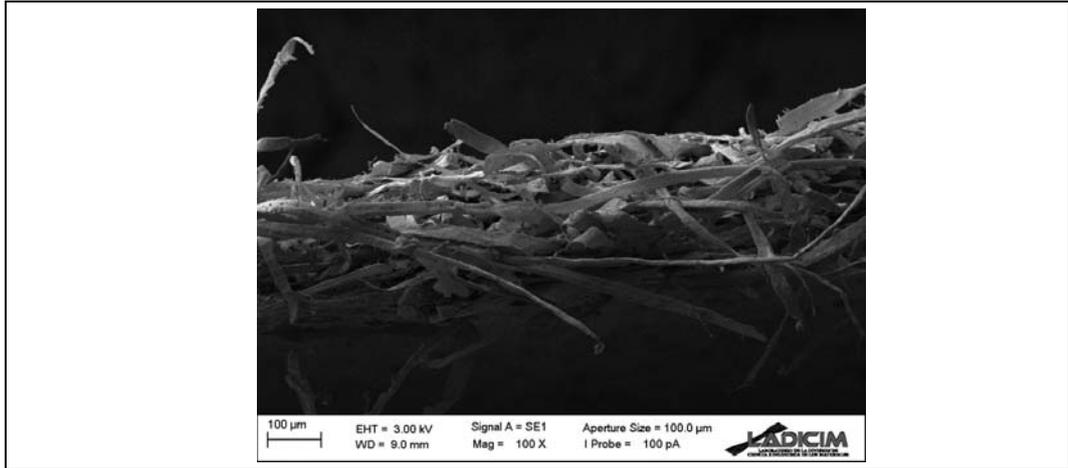


Fig. 12: Rupture section of the new Kraft paper

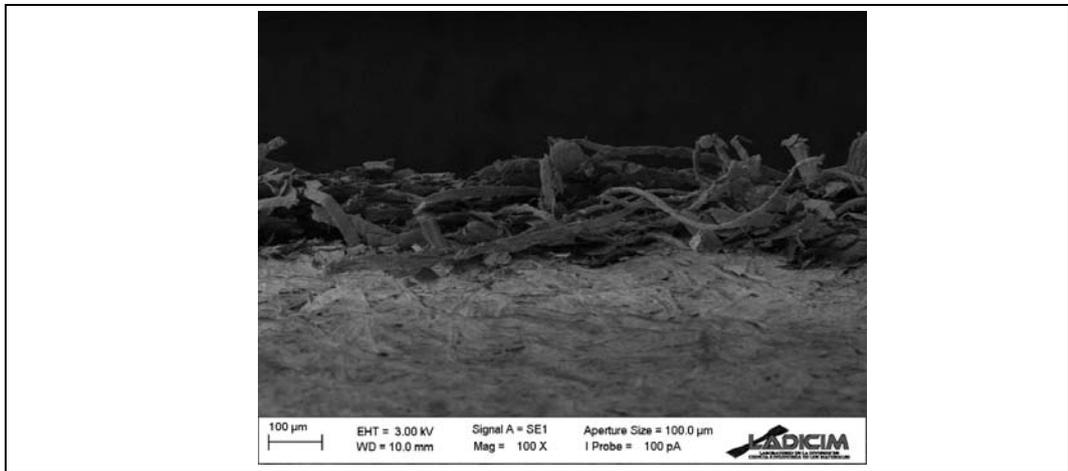


Fig. 13: Rupture section of Kraft after 72 h at 130°C

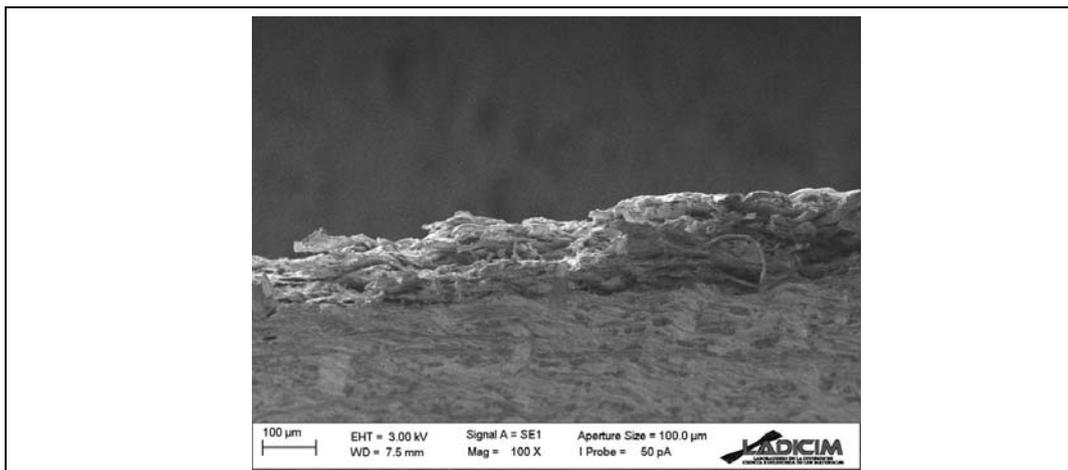


Fig. 14: Rupture section of Kraft after 168 h at 130°C

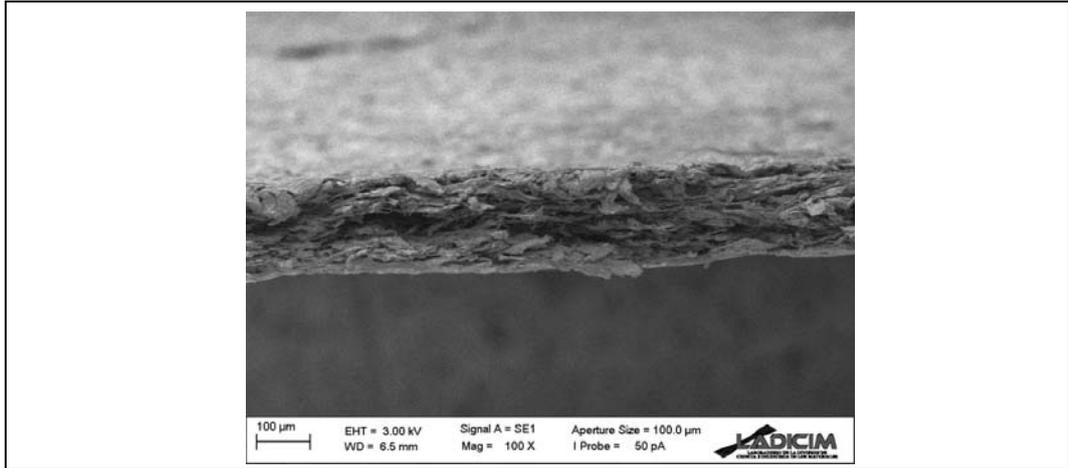


Fig. 15: Rupture section of Kraft after 260 h at 130°C

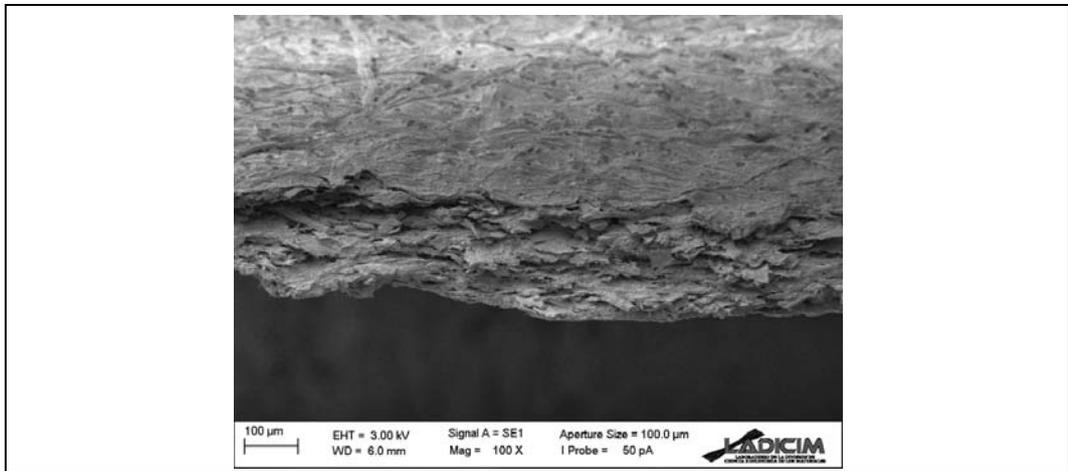


Fig. 16: Rupture section of Kraft after 425 h at 130°C

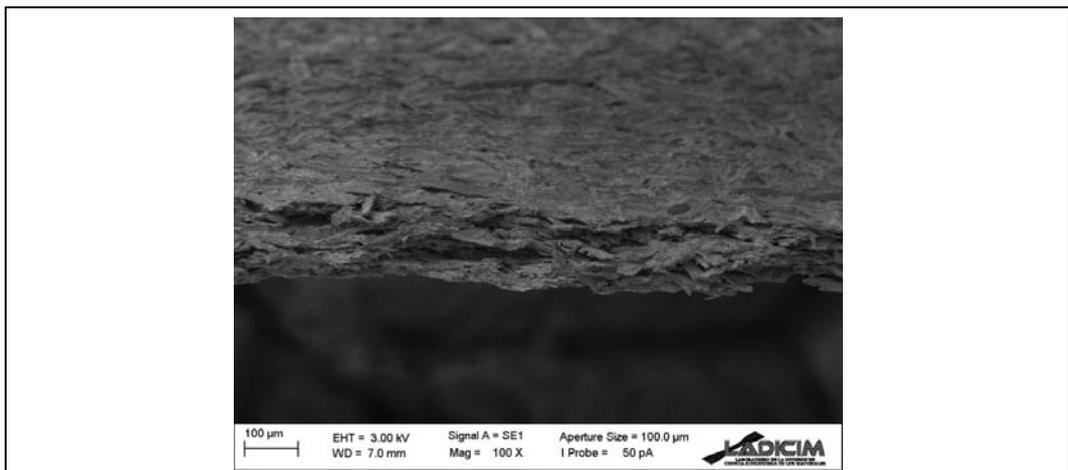


Fig. 17: Rupture section of Kraft after 735 h at 130°C

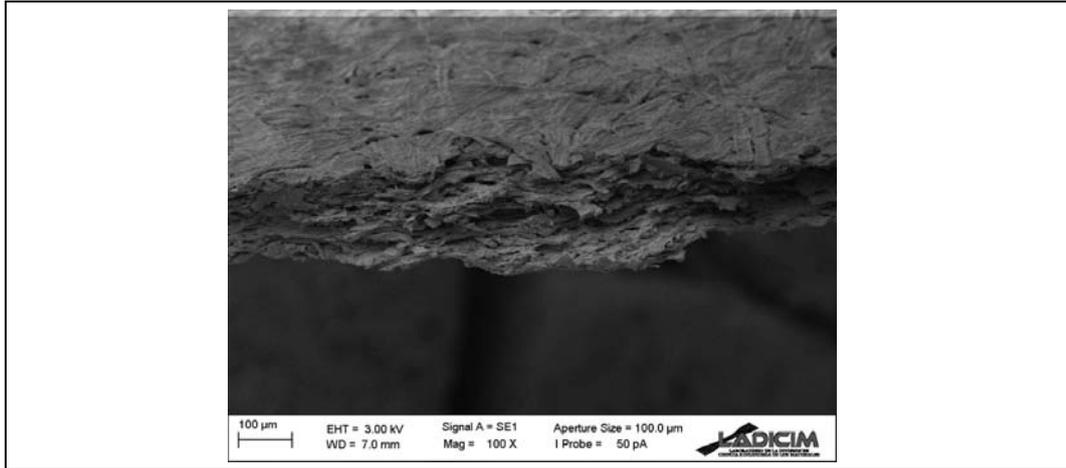


Fig. 18: Rupture section of Kraft after 1083 h at 130°C

CONCLUSIONS

A behaviour model based on DP has been defined to predict the lifespan of Kraft paper used in the dielectric system of power transformers. This model is a function of the time and the temperature at which the paper is aged.

Also, the loss of mechanical properties has been analyzed as the paper deterioration progresses, proving that the mechanical properties of the paper are already minimal before reaching the critical conditions established for the DP.

Therefore, it might be advisable to revise the critical values of properties such as DP and σ_R in order to establish when the end of life of dielectric paper has been achieved. Additionally, a fractography analysis should be carried out to include the results in new mathematical models that consider not only new critical values, but also values which mark the change of failure mode undergone by Kraft paper when this is used in power transformers as a dielectric solid.

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