Assessment of the effect of commercial vegetal oils on Kraft paper ageing through mechanical characterization

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ABSTRACT

Vegetal oils are considered a suitable substitute of mineral oil, widely used in power transformers as insulation liquid and cooling. Due to the fact that more and more power transformers containing this alternative insulating fluids are being constructed it is needed the development of mathematical aging models which help to predict transformer failures. The continued performance of power transformers depends on the condition of its paper insulation mainly. In this sense, this paper analyses Kraft paper degradation through the loss of its mechanical strength. Accelerated thermal ageing test of the paper in two different vegetal oils were carried out at three temperatures during diverse periods of time, in order to obtain information on the kinetics of the ageing degradation of the paper. The evolution of the mechanical properties 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.

Index Terms — **Kraft paper, power transformer, thermal ageing, tensile test, degree of polymerization, vegetal oil.**

1 INTRODUCTION

Power transformers play a critical role in transmission and distribution electrical networks [1-2]. During their operation these machines suffer thermal, electrical, mechanical and chemical stresses which degrade insulation system [3]. This insulation system usually consists of dielectric oil and paper. The functions of the liquid are provide electrical insulation and absorb and transmit the heat produced by energy losses to reduce the rise of temperature in the windings and cores. On the other hand, the dielectric solid provides both electrical and mechanical support in power transformers, its immersion in insulating oil increases its electrical insulation strength [4].

Kraft paper (90% cellulose, 6-7% lignin and 3-4% pentosans) [5] is one of the most widely used solid material due to economic factors and ease of manufacture, although other materials are being used [3, 6-7]. At the same time, the most widely used dielectric liquid in power transformers is mineral oil, which exhibits excellent dielectric and coolant behavior [8]. However, this oil has low flash and fire points, as well as, very low biodegradability. These shortcomings have led the search for new fluids [9]. Different works have concluded that vegetal oils exhibit a good dielectric and improved environmental performances [10-11]. Consequently, this new kind of dielectric oil might represent a suitable alternative to mineral oil if its interaction with solid insulation not accelerate the normal aging of the insulation system.

It has been found that the power transformer component related failure is placed in the insulation system, contributing to about 41% from the total failures [11]. During power transformers operation, both dielectric solid and liquid undergo continuous degradation [1, 12-13]. However, the main cause of transformers failure is mechanical damage of solid insulation, therefore the reliability of power transformers depends on the lifespan of dielectric paper, principally [5, 14-16]. Since the lifetime of a transformer depends fundamentally on the state of the solid insulation, a detailed assessment of dielectric paper condition is highly beneficial. Estimating the paper's life can be used by management planners to decide the most suitable moment to change a transformer [17-18]. Therefore, if the use of natural esters in more and more power transformers is desired, it is critical to evaluate the stability of the new insulations systems based on vegetal oils in comparison with the usual system in power transformers (mineral oil / Kraft paper).

There are several studies that have carried out thermal aging tests in laboratory to evaluate the behavior of different types of dielectric papers in insulation oils. Firstly, these works were focused on mineral oil [6-7, 19]. Nevertheless, since last two decades the resistance of dielectric papers which are aged in alternative insulation liquids, has started to be studied. For instance, McShane et al. [20] compared reaction rates of thermally upgraded and Kraft paper aged in natural ester and in mineral oil at 130, 150 and 170ºC. This assessment took into account tensile strength and degree of polymerization (DP). The behavior of thermally upgraded and Kraft paper was also studied by Abdelmalik [21] who aged these papers in mineral and palm kernel oil at 150ºC. In this work was evaluated the evolution of tensile strength, energy at maximum tensile stress and breakdown strength of the paper. The degradation of Kraft paper aged in natural ester at 120 and 150ºC was also studied by Widyanugraha et al. [22] through testing of tensile strength. Additionally, these authors measured dissolved gas in dielectric oil. The failure rate of transformer insulation systems made up of Kraft paper and mineral oil and two different vegetal oils was evaluated by Madavan and Balaraman [23]. They aged the insulation systems at 140ºC and tested dielectric strength, acidity, dissipation factor, resistivity and tensile strength. Raof et al. [4] analysed the potential of palm-based neopentyl glycol diester as dielectric fluid. They aged this liquid and mineral oil at 90, 110 and 130ºC to compare their behavior through acidity, viscosity, flashpoint and breakdown voltage. Moreover, they compared mechanical properties of Kraft paper aged in both fluids. All these works obtained a slower ageing rate of solid insulation in vegetal oils. On the other hand, Saruhashi et al. [24] carried out accelerated thermal ageing tests of aramid paper in silicone, natural and synthetic ester at 130 and 180ºC. They analysed color changes, total acid number, kinematic viscosity, dielectric breakdown voltage and tensile strength. It was concluded that mechanical resistance of paper remained unchanged during test time. These authors proposed tests of longer duration to obtain a more detailed study of paper degradation for future research.

Although there are various methods to evaluate the aging condition of the insulating paper [3], the degree of polymerization (DP) and the tensile strength are the most commonly used in the literature. 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, but also it has been shown that paper undergoes a decrease in its mechanical properties. For this reason, several authors have modeled the relationship between the variation of the tensile strength and the degree of polymerisation (DP) over time [25-26]. Another more recent studies [21, 27-30] have correlated the variation of the tensile strength with properties of the oil such as: dissolved gases, furans content [28], breakdown voltage, acidity and water content [27, 31].

As has been mentioned previously, the development of degradation models is essential to provide tools which can make predictions on when it is the most suitable moment to replace a power transformer. Although, there is an important amount of works that have defined kinetic models based on tensile strength, additional mechanical properties as energy consumed per unit volume or strain, which are closely related to the brittleness of the material, have not been taken into account.

Considering the importance that other mechanical parameters might have in the analysis of paper degradation, this work attempts to correlate the variations suffered by different mechanical properties (strength, Young's Modulus, yield stress, energy consumed, etc.) with the ageing suffered by Kraft paper in two commercial vegetal oils. The final aim is to establish whether data obtained from the stress-strain curve might be able to provide additional information which could provide a more accurate prediction of dielectric paper failure. Furthermore, a mathematical model has been defined based on the degree of polymerisation to predict the remaining life of Kraft paper aged in vegetal oils. 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 properties (DP, strength, Young's Modulus, yield stress, energy consumed…) supplies the more useful information and whether the end-of-life criteria of the dielectric material established until now (DP=200, 25% retained tensile strength [18] are the most suitable for guaranteeing the reliability of the solid insulation.

2 MATERIAL

This work aims to assess the degradation rate of Kraft paper immersed in two commercial vegetal oils (Table 1) taking into account paper anisotropy through mechanical analysis. The analysis of the Kraft paper (Table 2) degradation, it has been carried out through accelerated thermal aging tests.

In order to get insulation paper samples with different aging level, Kraft paper was cut into strips of 250 mm in length and 15 mm width. Due to paper anisotropy, these strips were cut with different fiber direction angles (longitudinal, transverse and 45º).

Table 1. Properties of vegetal oils analysed.

Property	Units	Standard	Vegetal oil 1 (V01)	Vegetal oil 2 (V02)
Viscosity, 0ºC	mm^2/s		232	275,9
Viscosity, 40ºC	mm^2/s	ISO 3104, ASTM D445	37	39,2
Viscosity, 100ºC	mm^2/s		8,3	8,5
Density, 20ºC	kg/dm^3	ISO 3675, ASTM D4052	0,92	0,91
Breakdown voltage (2.5 mm)	kV	IEC 60156	>75	65
Acidity	mg KOH/g	IEC 62021.3	$\leq$ 0,04	0,05
Dielectric dissipation factor (90ºC)		IEC 60247	0,05	0,03
Flash point	ºC	ISO 2592, ASTM D92	>315	330
Fire point	ºC	ISO 2592, ASTM D92	>350	362
Pour point	ºC	ISO 3016, ASTM D97	-31	-25
Water content	mg/kg	IEC 60814	50	150

Table 2. Kraft paper properties.

Property	Units	Standard	Value
Grammage	g/m^2		149.3
Thickness / 5 sheets	µm	IEC 60554-2	198
Apparent density	kg/m^3		754
Ash content	%		<0.6
Aqueous extract conductivity	mS/m		1.5
Dry breakdown strength in air	kV/mm		8.9

3 EXPERIMENTAL METHODOLOGY

3.1 THERMAL AGING

Trying to reduce the influence of paper moisture on the ageing, Kraft specimens were placed into a stainless steel vessel which has a volume of 1 liter and two connections, one for the vacuum pump and another for the vacuometer, as is shown in Figure 1.

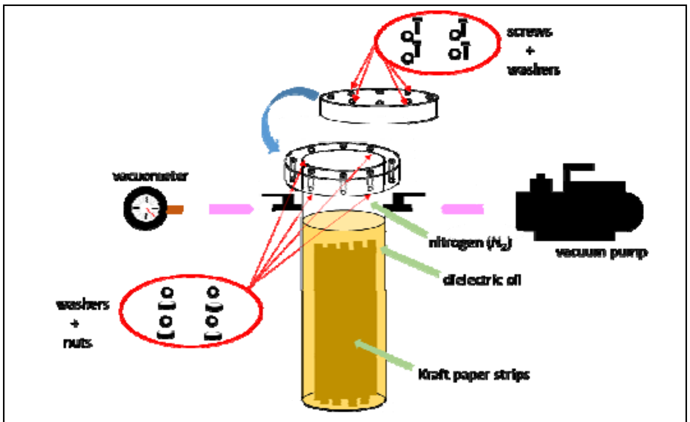

Figure 1. Stainless steel design test tube used for thermal ageing tests

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 before ageing tests, providing samples with a moisture content of 2%.

After that, paper strips impregnated in 750 ml of new oil (two commercial vegetal oils) into the vessel with a nitrogen headspace of 25% by volume. Finally, the vessel was put into an oven for thermal ageing at different temperatures: 110, 130 and 150°C. In this work, 7 groups of strips for Kraft paper were prepared, one group of new paper and the rest of thermal aged samples.

3.2 CHARACTERISATION OF CELLULOSE DEGRADATION TROUGH DP

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 20ºC 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 property the DP was obtained. For this, it is necessary to know the moisture of the sample, which was determined using Karl Fischer titration (IEC 60814).

3.3 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, E_R.

4 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. The last one offers a more detailed analysis of dielectric paper degradation because it allows compare the effect of fiber direction angles on mechanical resistance, which cannot be observed with DP measure.

4.1 Paper Ageing Model Based on DP

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 for both commercial vegetal oils that the DP decreases over time and this decrease becomes faster when the ageing temperature increases. The Kraft paper immersed in both vegetal oils shows a quite similar ageing rate.

Kraft paper degradation based on DP can be modeled as a function of time and temperature using a damage parameter D based on equation (1):

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

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.

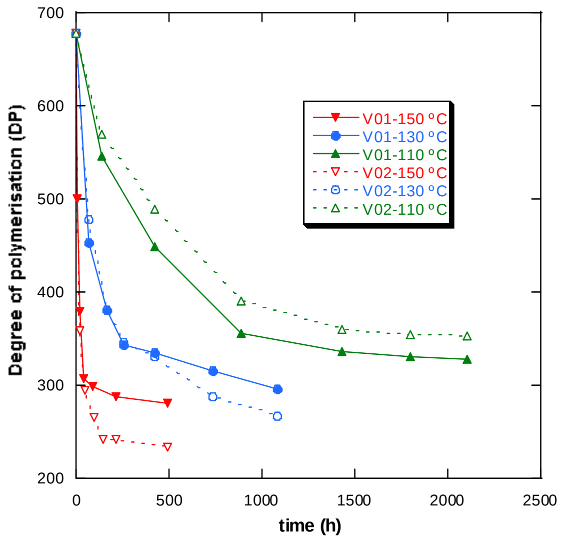

Figure 2. DP evolution of Kraft paper immersed in V01 and V02

This parameter of damage D, can be also expressed as a function of the t and T:

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

where the constant a depends on the ageing temperature and D_{max} is the maximum value allowed for the damage. D_{max} can change depending on the critical degree of polymerization (DP_c) used as the end-of-life criteria. Though a DP_c = 200 is often established as end-of-life criteria [18]. This work has considered different DP_c to obtain a better correlation with experimental data. Table 3 gathers the correlation coefficients obtained for different values of DP_c to estimate D_{max} used in equation (2), for the two vegetal oils.

Table 3. Effect of DPc on damage estimation.

DPc	Dmax	T (ºC)	R^2	a
Vegetal oil 1 (V01)				
200	0.705	110	0.910	$1.005 \cdot 10^{-3}$
		130	0.832	$4.727 \cdot 10^{-3}$
		150	0.899	$4.520 \cdot 10^{-2}$
250	0.631	110	0.948	$1.433 \cdot 10^{-3}$
		130	0.933	$6.932 \cdot 10^{-3}$
		150	0.980	$6.088 \cdot 10^{-2}$
300	0.556	110	0.988	$1.420 \cdot 10^{-3}$
		130	0.998	$1.046 \cdot 10^{-2}$
		150	0.988	$7.704 \cdot 10^{-2}$
Vegetal oil 2 (V02)				
DPc	Dmax	T (ºC)	R^2	a
200	0.705	110	0.925	$7.893 \cdot 10^{-3}$
		130	0.917	$4.670 \cdot 10^{-3}$
		150	0.971	$3.860 \cdot 10^{-2}$
250	0.631	110	0.950	$1.030 \cdot 10^{-3}$
		130	0.973	$6.560 \cdot 10^{-3}$
		150	0.997	$5.430 \cdot 10^{-2}$
300	0.556	110	0.979	$1.495 \cdot 10^{-3}$
		130	0.991	$9.436 \cdot 10^{-3}$
		150	0.958	$8.111 \cdot 10^{-2}$

It can be observed that DPc = 300 for both vegetal oils allows to obtain a more accurate model to predict the damage suffered by solid insulation during thermal aging tests in the laboratory.

The constant a, equation (2), can be expressed by means of an exponential law equation (3) which depends on the ageing temperature.

$$a = A \cdot \exp(B \cdot T) \qquad (3)$$

Table 4. Effect of DPc on exponential law equation (3).

		Vegetal oil 1 (V01)		Vegetal oil 2 (V02)	
DP_c	D_{max}	A	B	A	B
200	0.705	$2.543 \cdot 10^{-8}$	$9.515 \cdot 10^{-2}$	$1.692 \cdot 10^{-8}$	$9.722 \cdot 10^{-2}$
250	0.631	$4.317 \cdot 10^{-8}$	$9.373 \cdot 10^{-2}$	$1.829 \cdot 10^{-8}$	$9.906 \cdot 10^{-2}$
300	0.556	$1.067 \cdot 10^{-8}$	$8.971 \cdot 10^{-2}$	$2.415 \cdot 10^{-8}$	$9.984 \cdot 10^{-2}$

Consequently, the DP_i can be expressed by means of equation (4) as a function of the time and the temperature.

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

This equation make possible to obtain the theoretical behaviour of Kraft paper in these two vegetal oils, for other temperatures, including those typical of the operation of power transformers (60-90ºC).

Figure 3 shows the evolution of the damage parameter, D, with t for three ageing temperatures. This figure shows the good correlation between the experimental data and the adjustment performed according to equation (2) for vegetal oil 1 (V01) and $DP_c = 300$.

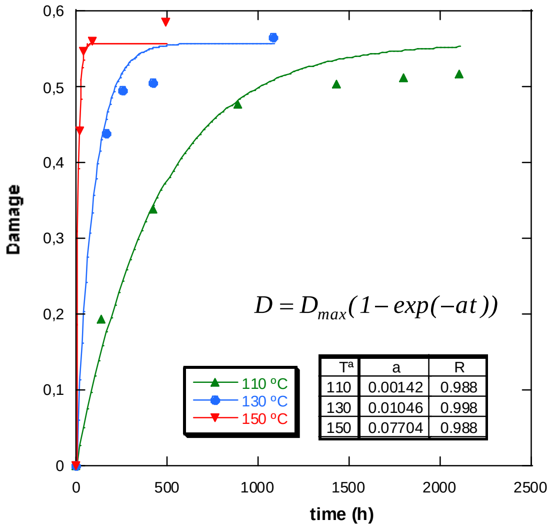

Figure 3. Evolution of the damage, D, in function of the t and the T for V01 and DPc = 300

The equation (5) obtained from equation (2), can be used to calculate the time, t, needed to obtain a value of DP_c established as D_i, depending on the operating temperature of the transformer. It can be verified that when the ageing rate increases (higher temperature), the time decreases logarithmically.

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

4.2 PAPER AGEING MODEL BASED ON TENSILE CHARACTERISATION

When the mechanical behaviour of the original paper is analysed as a function of the fibre direction angle, gathered in Figure 4, 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 45º are intermediate and are discarded for the rest of this study.

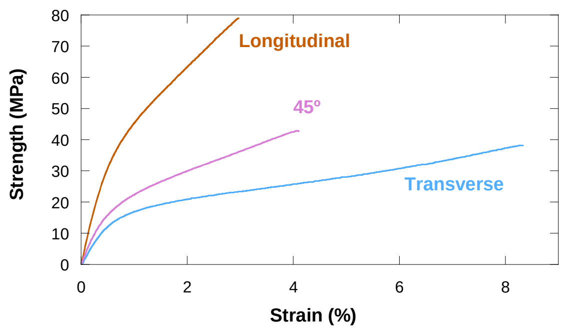

Figure 4. Anisotropy of Kraft paper

The assessment of the evolution of the strength-strain curves, obtained for different ageing times for the three ageing temperatures considered in this study, has verified that the Young's Modulus can hardly provide any information on the Kraft paper deterioration, since it is quite similar in all the curves. 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 those that clearly depend on the paper degradation.

Figures 5 and 6 show 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. Additionally, it can be observed in these two figures 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 when ageing temperatures are higher, but with relatively poor values. It can be verified that the fibres in the rupture have undergone significant strain when the Kraft paper is new. However, once the paper has been aged this strain is reduced, even before 100 h when ageing temperature is 150ºC, which produce a paper rupture quite more fragile. This fall 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. This loss of properties is observed in a similar way when fibre orientation is longitudinal and transverse.

Regarding the energy consumed in the rupture, E_R, which is a combination of strength and strain, time and temperature have a greater effect, Figures 7 and 8.

Kraft paper degradation based on tensile characterisation can be modeled as a function of time and temperature using a damage parameter D, in a similar way to how it has been down for DP, equation (2). D_{max} can change depending on the critical rupture strength (σ_{Rc}) used as the end-of-life criteria, Table 5.

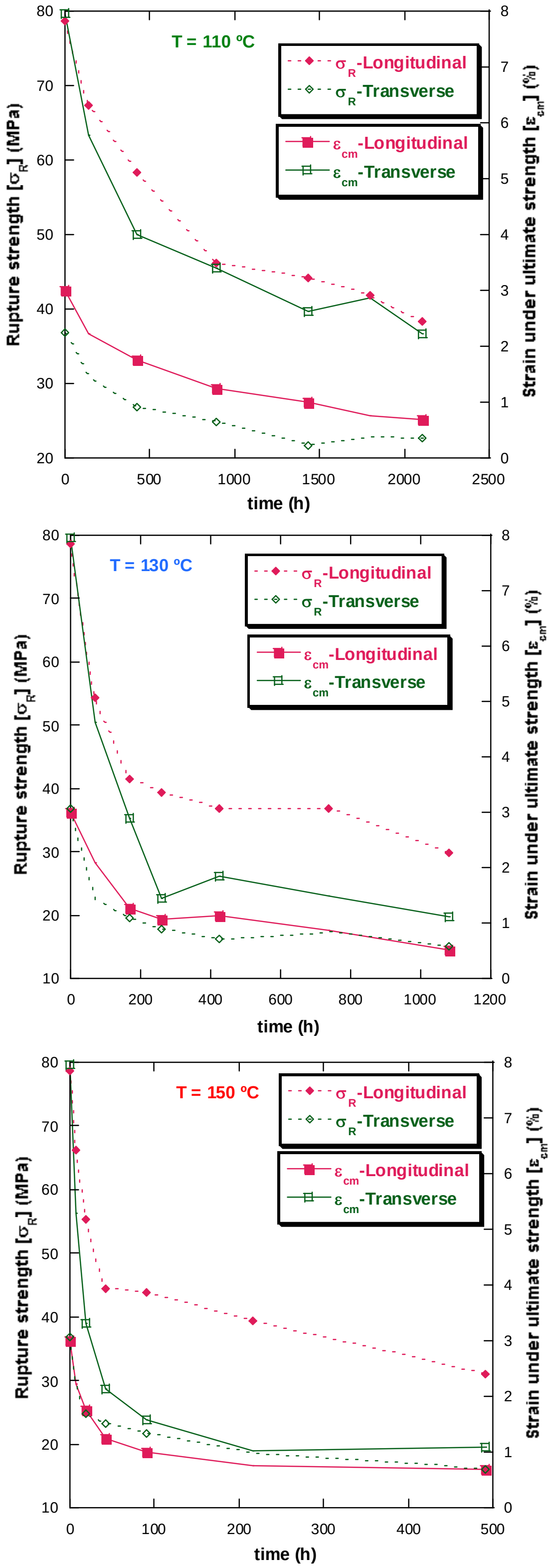

Figure 5. σ_R and ε_{cm} as a function of the ageing t for V01

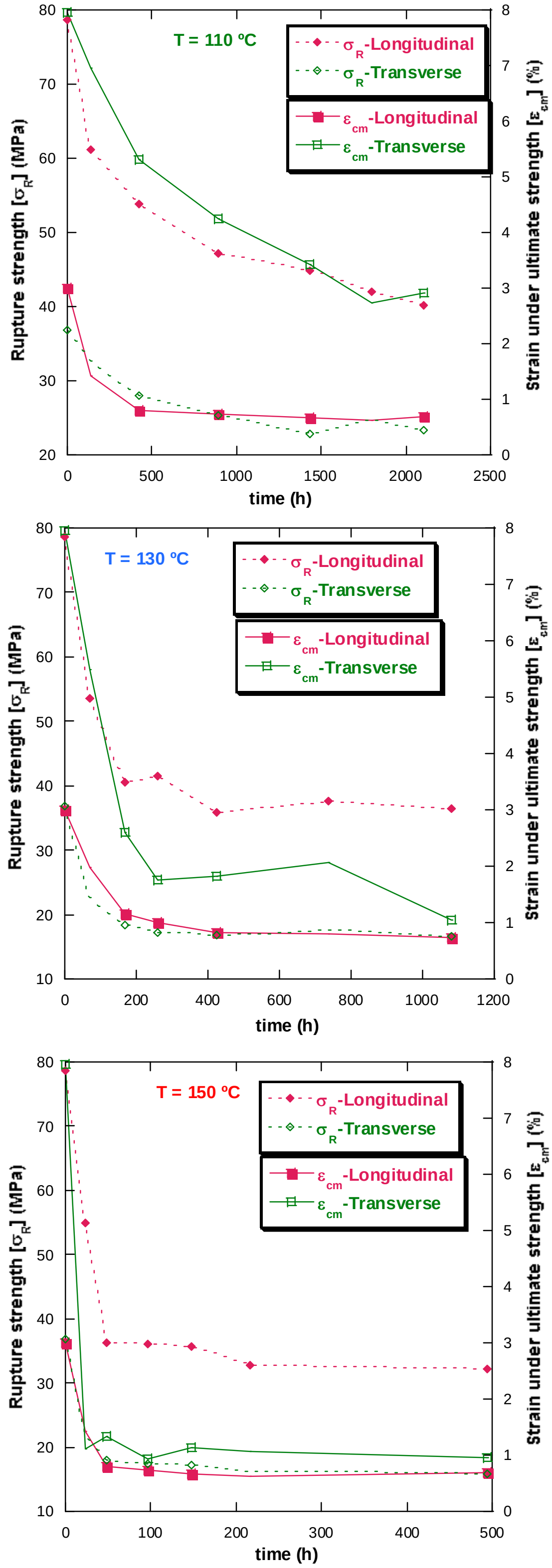

Figure 6. σ_R and ε_{cm} as a function of the ageing t for V02

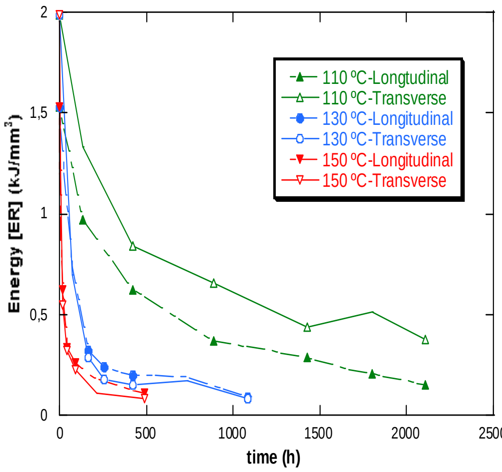

Figure 7. E_R as a function of time and temperature for V01

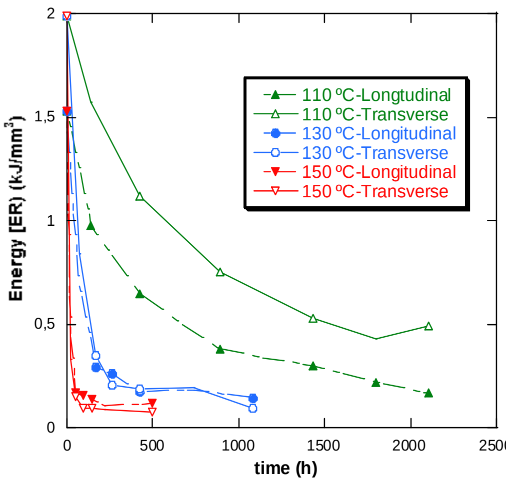

Figure 8. E_R as a function of time and temperature for V02

Table 5. Effect of σ_{Rc} on damage estimation for transverse fibre orientation.

σ_{Rc}	Dmax	T (ºC)	R^2	a
Vegetal oil 1 (V01)				
0.45* σ_{R0}	0.55	110	0.877	$9.280*10^{-4}$
		130	0.991	$1.510*10^{-2}$
		150	0.939	$3.420*10^{-2}$
0.50* σ_{R0}	0.50	110	0.912	$1.230*10^{-3}$
		130	0.973	$2.050*10^{-2}$
		150	0.961	$4.620*10^{-2}$
0.55* σ_{R0}	0.45	110	0.954	$1.830*10^{-3}$
		130	0.909	$2.860*10^{-2}$
		150	0.988	$6.190*10^{-2}$
Vegetal oil 2 (V02)				
σ_{Rc}	Dmax	T (ºC)	R^2	a
0.45* σ_{R0}	0.55	110	0.882	$7.430*10^{-4}$
		130	0.997	$1.630*10^{-2}$
		150	0.997	$5.640*10^{-2}$
0.50* σ_{R0}	0.50	110	0.907	$9.310*10^{-4}$
		130	0.987	$2.170*10^{-2}$
		150	0.977	$7.600*10^{-2}$
0.55* σ_{R0}	0.45	110	0.939	$1.270*10^{-3}$
		130	0.927	$2.980*10^{-2}$
		150	0.908	$1.130*10^{-1}$

5 CONCLUSION

The deterioration data of Kraft paper immersed in two commercial vegetal oils during accelerated thermal aging tests, have been used to develop models based on DP and tensile characterization that can estimate ageing rate of Kraft paper in other conditions of time and temperature. These models based on laboratory data have showed that the end-of-life criteria of the dielectric material established until now, might have to be revised in order to obtain more accurate models. The results obtained for both vegetal oils are quite similar, therefore the proposed models should be used to estimate the ageing rate of Kraft paper immersed in other vegetal liquids in order to prove the suitability of the proposed models.

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