Fracture Toughness As An Alternative Approach To Quantify The Ageing of Insulation Paper In Oil

Cristina Fernández-Diego (fdezdiegoc@unican.es)
University of Cantabria: Universidad de Cantabria
Isidro Alfonso Carrascal
University of Cantabria: Universidad de Cantabria
Alfredo Ortiz
University of Cantabria: Universidad de Cantabria
Inmaculada Fernández
University of Cantabria: Universidad de Cantabria
Diego Ferreño
University of Cantabria: Universidad de Cantabria
Soraya Diego
University of Cantabria: Universidad de Cantabria
José Antonio Casado
University of Cantabria: Universidad de Cantabria

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Abstract

Oil-immersed transformers use paper and oil as insulation system which degrades slowly during the operation of these machines. The fast-developing electric power industry demands superior performance of electrical insulation materials which has led to the development of new materials whose measurement of the degree of polymerization has found some practical difficulties. Moreover, the increasing interest in synthetic dielectric materials replacing cellulose materials requires the use of alternative methods to the degree of polymerization to quantify the degradation of insulation solids over time. In this sense, this paper proposes the possibility of analyzing paper degradation through fracture toughness. An accelerated thermal ageing of Kraft paper in mineral oil was carried out at 130ºC during different periods of time, to obtain information on the kinetics of the ageing degradation of the paper. Double-edged notched specimens were tested in tension to study their fracture toughness. The evolution of the load-displacement curves obtained for different ageing times at the ageing temperature of 130°C was utilized to the determination of the stress intensity factor. Furthermore, different kinetic models based on this stress intensity factor were applied to relate its evolution over time as a function of the temperature. Finally, the correlation between the DP and stress intensity factor, which depends on the fiber angle, was also defined.

Highlights

- Fracture toughness provides information about the effect of paper anisotropy.
- The degree of polymerization and fracture toughness display a good agreement.
- Fracture toughness allows the thermal ageing to be evaluated over time.

Introduction

Transformers are long run devices known as key parts of an electrical power system (Maharana et al. 2018; Karthik and Narmadhai 2020; Suwarno and Ritonga 2020). Nowadays, oil-immersed transformers, whose insulation system is composed of an insulating fluid (mineral oil, natural or synthetic ester) and a conductor insulation (Kraft, Crépe, DDP, Nomex®…) are widely used worldwide (Chen et al. 2020; Garelli et al. 2021). Cellulose insulants are the main solid insulator for the winding conductors, inter-winding, intercore and winding-to-earth insulation (Levchik et al. 1998; Rao and Jarial 2019) due to its excellent insulation performance and mechanical properties at elevated temperatures (Medina et al. 2017; Jusner et al. 2021).

During transformers' operation their paper and oil insulations are affected by many variables (temperature, moisture, oxygen, mechanical and electric stresses), provoking their degradation and affecting their dielectric, mechanical and chemical properties (Li et al. 2020). Although the degradation suffered by oil can be resolved through its treatment (filtration, dehumidification, purification) or replacement, paper deterioration can only be repaired by means of paper replacement which is critical for

economic reasons (Łojewski et al. 2010; Medina et al. 2017). Therefore, the lifespan of a transformer is considered to be mainly defined by the insulation paper condition (Łojewski et al. 2010; Chen et al. 2020).

When insulation paper ages, depolymerization of the cellulose chain takes place which results in a decrease of its degree of polymerization (average number of glucose units per cellulose chain) and mechanical strength. Therefore, the insulation material becomes more brittle and carbonaceous and reduces its capability to withstand thermal, mechanical and electric stresses (Oommen and Prevost 2006a part I; 2006b part II; Vänskä et al. 2014).

The condition and remaining life of the winding insulating paper can be evaluated through indirect measurements (furaldeydes, gases, methanol or ethanol dissolved in the oil, energy dispersive spectroscopy, Fourier-transform infrared spectroscopy, differential thermal analysis…) and direct methods (degree of polymerization (DP), mechanical properties) (Arroyo et al. 2015; Saldivar-Guerrero et al. 2016; Suwarno and Ritonga 2020). These direct methods have been used to evaluate the compatibility of cellulose and new liquids, degradation, damage and ageing in different works (Hill et al. 1995; Li et al. 2016; Hosoya et al. 2018; Wang et al. 2019; Faiz et al. 2020). The end-of-life criteria applied to insulation paper is based on either a value of 200 for DP or retained tensile strength (20%) as the decisive factor to determine the material lifespan. However, the need of high performance electrical insulation materials (Prevost and Oommen 2006; Chen et al. 2019; Jindal and Singh 2020; Xie et al. 2020), as well as new industrial drying processes have produced new dielectric materials in which the determination of DP has been found problematic (Lukic et al. 2021). Since the DP is a specific property of cellulose products (Marek and Szewczyk 2017), mechanical properties seem to be more suitable to quantify the insulation deterioration, working universally for almost all solid materials.

The most commonly used mechanical property for assessing the condition of the insulation paper is the tensile strength (Hill et al. 1995) which has been found that decreases proportionally to the DP (Medina et al. 2017). Experimental results on the thermal ageing of Kraft paper in different insulating liquids (mineral oil, natural ester and gas to liquid oil) using the DP and the tensile strength were reported by Suwarno and Ritonga (Suwarno and Ritonga 2020). The analysis of tensile strength and DP was applied by Maharana et al. (Maharana et al. 2018) to measure the mechanical strength and the intensity of chemical degradation of Kraft paper too. Their experiments on thermal ageing were performed for two different insulating oils (mineral oil and a nanofluid based on exfoliated hexagonal boron nitride). These authors found that the DP and the mechanical strength of the oil impregnated Kraft paper were proportional to each other. A linear correlation between tensile strength and methanol was also concluded by Arroyo et al. (Arroyo et al. 2015, 2017) who studied two paper/oil systems (standard wood Kraft paper and a thermally-upgraded Kraft paper) under ageing conditions at a large range of temperatures (150-190^{o}C). They fitted their experimental data by means of a kinetic model, defined by Calvini, using the Arrhenius equation to simulate the evolution of the mechanical properties' during thermal ageing. The applicability of methanol as an ageing marker was also evaluated by Matharage at al. (Matharage et al. 2018), who studied the thermal ageing of an insulation paper immersed in synthetic ester at 80, 100 and 120^{o}C using the tensile index. Furthermore, tensile strength has been used to measure the effect of accelerated

thermal ageing of thermally upgraded Kraft and cellulose/aramid papers (Nomex 410 and 910) for improving the understanding of ageing in the mechanical properties of these relatively new materials (aramid papers) (Ranga and Chandel 2019; Arroyo et al. 2020). Arroyo-Fernández et al. (Arroyo et al. 2020) obtained a quasi-linear or master curve between the DP and the tensile strength for the cellulose-based paper. Moreover, they used the reduction in elongation over time to determine the state of solid insulation in power transformers, which demonstrated that this mechanical property can also be a suitable monitoring tool.

Even though there are different studies in which the suitability of mechanical properties to measure the deterioration undergone by a solid insulation has been demonstrated, to the best of these authors' knowledge, there are no contributions based on the use of fracture toughness to quantify the insulation solid deterioration and to predict its available lifespan. Fracture toughness represents the ability of a cracked material to resist fracture. A reduced fracture toughness indicates that a material is susceptible of undergoing brittle fracture, while a high fracture toughness is associated with a ductile material. Considering the significance of this novel approach, this paper is aimed to correlating the variations experienced by the fracture toughness with the degree of ageing of a Kraft paper. In addition, four kinetic models obtained from the bibliography have been applied to envisage the remaining lifespan of the Kraft paper using the stress intensity factor variation and their predictions were compared with those obtained from a classical model based on the DP and the load-displacement curves.

Materials And Thermal Ageing

Material

For this study, the Kraft paper, whose properties are showed in Table 1, was selected. This paper was cut into strips with a length of 64 mm and a width of 15 mm. Due to the anisotropy of the paper, these strips were used to manufacture specimens with the two orientations of the paper: machine direction (MD) and cross machine (CD).

This work studies the deterioration of Kraft paper immersed in mineral oil (Table 2) during thermal ageing at 130^{o}C. The degradation rate is evaluated using the degree of polymerization, through tensile tests (ultimate tensile strength, σ_R, strain under ultimate strength, ε_{cm}, and energy per unit volume, E_R) and fracture toughness tests.

Table 1. Kraft paper properties.

Property	Units	Value
Grammage	g/m^2	149.3
Thickness / 5 sheets	Mm	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

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

Thermal ageing

Kraft strips were placed into a stainless-steel vessel (Fig. 1) at pressure of 1 mbar and temperature of 100°C for 24 hours in order to reduce its moisture content to 2%.

The dielectric mineral oil (750 ml) was added to the vessel, and this was filled with 25% of its volume with nitrogen. Subsequently, the vessel was placed inside an air-forced oven and the temperature was maintained at 130°C during the thermal ageing. The paper strips and oil were removed at different times (Table 3) for analysis. After the specific period of thermal ageing, six groups of paper samples with different deterioration levels were taken to characterize their properties (DP, ultimate tensile strength, strain under ultimate strength, energy consumed per unit volume and fracture toughness).

Table 3. Ageing time.

Temperature	Ageing time (h)						
	Samples						
	0	1	2	3	4	5	6
130ºC	0	72	168	261	425	736	1083

Traditional characterization of cellulose degradation

Kraft paper (90% cellulose, 6-7% hemicellulose and 3-4% lignin) used for covering conductors in transformers is often characterized through the DP which represents the polymer chain average length of the cellulose (Bandara et al. 2016; Saldivar-Guerrero et al. 2016). Cellulose is a linear polymer made of the glucoside bond of the beta-D glucose monomer through the 1,4-glycoside bond (Wang et al. 2018). The thermal stress suffered by Kraft paper during thermal ageing results in a decrease of the DP (Vänskä et al. 2014). Consequently, the DP is recognized as an objective detection method to measure the rate of degradation. In this study, the DP of each group of specimens was measured following the viscometric method of the standard ASTM D4243. The viscosity-average DP was obtained based on measurements at 20ºC using an automatic viscometer equipped with a two-sphere Ubbelohde tube. Each specimen was first de-oiled using hexane. Subsequently, the milled paper was dissolved in a solution (22.5 ml of deionised water and 22.5 ml of 1M solution of bis(ethylenediamine) copper (II) hydroxide). After dissolution of the paper in the solution through magnetic stirring, its specific viscosity was determined. Then, the intrinsic viscosity of the solution was deduced, and then the DP was obtained. The moisture of the sample, which is necessary for the previous calculations, was determined using the Karl Fischer titration.

Tensile tests were conducted using a servo hydraulic universal machine with an axial load cell of ± 1 kN capacity, an actuator of ± 50 mm of dynamic stroke and equipped with pneumatic flat grips. The tensile tests were carried out according to ISO 1924-2 2009 and the parameters obtained in the test were the yield stress, σ_y, tensile strength, σ_R, strain under ultimate strength, ε_{cm}, and energy per unit volume at the tensile strength, E_R.

Both DP and tensile strength have been accepted to be equally important criteria to assess the deterioration on insulation papers (IEEE 2018). However, since DP is specific to cellulose products and tensile strength works for almost all solid materials used in transformers' insulation systems (Marek and Szewczyk 2017), the second one might be considered more suitable to establish a lifespan criteria.

Fracture toughness

Fracture toughness tests are carried out to quantify the resistance of a material to failure by cracking (Anderson 2005). The stress intensity factor K_I measures the stress state near the tip of a crack

or notch caused by a remote load or residual stresses. The constraint conditions at the tip of a crack are affected by the thickness of the component: thin components (like the pieces of paper characterized in this research) display pale stress conditions and thick components plane strain conditions. Plane strain conditions give the lowest fracture toughness value which is a material property. The critical value of stress intensity factor in tensile (mode I) loading measured under plane strain conditions is known as the plane strain fracture toughness, denoted K_{Ic}. When a test fails to meet the thickness and other test requirements that are in place to ensure plane strain conditions, the fracture toughness value produced is denoted K_c. The fracture process zone in a piece of paper corresponds to the region just ahead of the crack tip, where the fiber breakage and bond breakage concentrate when a cracked specimen is strained (Mai et al. 1995). Fracture toughness test were conducted following the K_{Ic} procedure of the standard ASTM E1820-01.

Double-edge notched specimens (Fig. 2) with different notch sizes, were tested in tension. Following the recommendations of previous authors (Mai et al. 1995; Chen et al. 2016; Mao et al. 2017) the ligaments of the specimens in this research are in the range 6 mm $\leq b_0 \leq$ 8 mm, with specimen width (2W) of 15 mm. Each specimen's ligament was measured using an optical device coupled to a three-dimensional coordinate measuring machine (TESA MICRO-HITE 3D). Fracture toughness tests were carried out at room temperature a universal servo hydraulic test machine with an axial load cell of ± 1 kN capacity, an actuator of ± 50 mm of dynamic stroke and equipped with pneumatic flat grips. The distance between the clamps was 60 mm and the crosshead speed to obtain the load-displacement curves was 6.67 mm/min.

The procedure to obtain K_{Ic} established in the ASTM E 1820-01 standard requires the previous calculation of a conditional result, K_Q which has to be obtained using the expression of the stress intensity factor provided by the standard introducing the critical load P_Q obtained through the procedure depicted in Fig. 3: an auxiliary secant line with a slope.

$$\left(\frac{Load}{Displacement}\right)_5 = 0.95 \cdot \left(\frac{Load}{Displacement}\right)_0 \quad (1)$$

is constructed, $(Load/displacement)_0$ being the slope of the tangent OA to the initial part of the curve. The intersection between the experimental curve and the auxiliary secant line determines $P_5 = P_Q$. K_Q in deep, double-edged, notched specimens has been obtained using equation (2):

$$K_Q = \frac{P_Q}{B \cdot \sqrt{W}} \cdot f\left(\frac{a}{W}\right) \quad (2)$$

where f(a/w) is a geometric factor given by (Anderson 2005):

$$f\left(\frac{a}{W}\right)=\frac{\sqrt{\frac{\pi\cdot a}{2\cdot W}}}{\sqrt{1-\frac{a}{W}}}\cdot\left[1.122-0.561\cdot\left(\frac{a}{W}\right)-0.205\cdot\left(\frac{a}{W}\right)^{2}+0.471\cdot\left(\frac{a}{W}\right)^{3}+0.190\cdot\left(\frac{a}{W}\right)^{4}\right] \quad (3)$$

The following conditions must be fulfilled to consider K_Q as a geometry-independent measure of fracture toughness:

- The crack length must be in the range $0.45 \leq a/W \leq 0.55$.
- The ratio (Pmax/PQ), where Pmax is the maximum load of the test, must be less or equal than 1.10.
- The thickness and length of the initial ligament must be higher than

$$2.5\cdot\left(\frac{K_Q}{\sigma_Y}\right)^{2} \quad (4)$$

where σ_y is the 0.2% offset yield strength in tension.

It the test results fails to meet at least one of these three qualification requirements then K_Q is not equal to K_{Ic} (the linear elastic, plane-strain fracture toughness) but it remains as a valid characterization of the material fracture toughness for the geometric characteristics (thickness) of the specimen.

Analysis And Results

The degradation of Kraft paper in mineral oil has been characterized using the following properties: DP, tensile strength, σ_R, strain under ultimate strength, ε_{cm}, and energy per unit volume, E_R.

Degree of polymerization

Figure 4 shows the evolution of the DP as a function of time. As can be seen, most of the reduction is concentrated in the first 200 hours (the DP is reduced below 50% of its initial value). According to Bandara et al. (Bandara et al. 2016), this drastic drop is the result of the thermal stress generated at high temperature which breaks the bonding of the cellulose chains. In addition, it is considered that most of the water in cellulose is in the amorphous region which makes this region of cellulose deteriorates more quickly than the crystalline one.

Different kinetic models based on the DP have been proposed in the literature (Ekenstam 1936; Emsley et al. 1997; Calvini and Gorassini 2006; Carrascal et al. 2018). These mathematical expressions relate the evolution of the DP over time as a function of the temperature. Once an end-of-life criterion is established, these models allow the remaining lifetime of the insulation paper to be estimated.

The kinetic parameters from the Calvini and Carrascal models (Calvini and Gorassini 2006; Carrascal et al. 2018) have been obtained considering a leveling-off degree of polymerization (LODP) or critical degree of polymerisation of DPc = 200 because this is often used as the end-of-life criterion (Lundgaard et al. 2007). As can be observed (Fig. 6), the model proposed by Emsley et al. (Emsley et al. 1997) provides the highest coefficient of determination R2 after the fitting of the free parameters.

Although the DP has proven to be an efficient tool to measure the loss of mechanical strength suffered by cellulosic materials such as the Kraft paper, it has also found some practical difficulties with its determination for some new papers after their drying (Lukic et al. 2021). Furthermore, the increasing interest in synthetic dielectric materials replacing cellulosic insulation (Prevost and Oommen 2006) enhances the establishment of alternative methods to DP to quantify the deterioration of insulation solids accurately. This role can be played by the properties obtained from tensile and fracture toughness tests, described hereafter.

Tensile tests

Firstly, it has been verified a strong anisotropy when the mechanical behavior of the original paper was analyzed as a function of the fiber direction angle (MD and CD). The ultimate tensile strength in MD is two times the one measured in CD, whereas the strain is half.

The correlation between a series of mechanical properties (ultimate tensile strength, σ_R, strain under ultimate strength, ε_{cm}, and energy per unit volume, E_R), and the DP has been used as a proxy to assess the reliability of those properties to characterize the ageing of the Kraft paper previously aged at 130^{o}C in the laboratory.

Figures 6 and 7 show the correlation between the DP and these mechanical properties. Both σ_R and ε_{cm} display a suitable linear correlation whereas the relation between E_R and the DP is sensibly exponential. Table 4 summarizes these mathematical relations and the correlation obtained for both fiber angle directions (MD and CD).

Table 4. Mechanical properties vs DP of Kraft paper aged at 130^{o}C

	Mathematical correlation	R^2
Cross direction (CD)		
σ_R	$DP = 16.25*\sigma_R + 91.70$	0.9663
ε_{cm}	$DP = 59.48*\varepsilon_{cm} + 215.93$	0.9684
E_R	$DP = 93.56*\ln(E_R) + 580.88$	0.9562
Machine direction (MD)		
σ_R	$DP = 7.16*\sigma_R + 127.45$	0.9927
ε_{cm}	$DP = 167.57*\varepsilon_{cm} + 197.48$	0.9761
E_R	$DP = 100.19*\ln(E_R) + 599.45$	0.9710

Stress intensity factor

Once the high correlation existing between the DP and some mechanical properties (σ_R, ε_{cm} and E_R) had been verified, the following phase was to study whether fracture toughness can be applied as an alternative tool to quantify the deterioration suffered by the insulating materials used in the construction of power transformers.

Figures 8 and 9 show the evolution of the load-displacement curves obtained for different ageing times at the ageing temperature of 130°C for MD and CD angle fiber.

It can be verified in Fig. 8 that the parameters of load and displacement clearly depend on the paper degradation. The rupture load and the displacement under this load are considerably affected by time. Similar behavior of properties loss was observed when fiber orientation was CD (Fig. 9).

The specimens analyzed in this work have not fulfilled the requirements explained previously to consider K_Q as a geometry-independent measure of fracture toughness, consequently K_Q is not equal to K_{Ic} but it is a valid characterization of the material fracture toughness for the thickness of the specimen.

The stress intensity factor (K_Q), whose evolution during thermal ageing is represented in Fig. 10, has been obtained using the load-displacement curves. It can be verified the ability of this mechanical parameter to quantify the influence of ageing on the material fracture strength. The K_Q is reduced below 32% of its initial value in the first 200 hours. Subsequently, this parameter continues reducing its value below 13% of its initial value although it requires more than 800 hours to achieve its final deterioration state.

The evolution of fracture strength was also verified through the study of rupture section of Kraft strips using scanning electron microcopy (SEM). Paper strips were viewed using a Carl Zeiss-EVO MA 15

microscope, which uses as its electrons source a Lanthanum hexaboride filament with different detectors to distinguish secondary and backscattered electrons and characteristic X-rays (Oxford Instruments).

In the fractographies (Fig. 11), it can be verified that the fibres in the rupture section have undergone significant strain when the Kraft paper is new and at the beginning of the ageing (73 hours of ageing) when the Kraft paper has suffered low deterioration (DP > 460). It can be appreciated the huge strain suffered by insulation solid. In new and low-degraded paper specimens, the fibers undergone significant strain because they have sufficient strength to withstand the load applied, being the loosening of the interface between matrix and fiber the reason why a greater displacement in the material is achieved. During thermal ageing, the fibers loss their mechanical resistance and then the failure occurs in the same plane than hemicellulose and lignin matrix, obtaining very low loads which are comparable with the matrix ones when resistance of insulation solid is considerable reduced (DP is close to 200). The fibres in the material have no strain which generates a fragile rupture. This change in the failure mode is critical because the crack propagation and final fracture in a fragile material takes place instantly which raise the partial discharges and short circuits occurrences.

Since K_Q has been demonstrated to be a suitable mechanical parameter to quantify the influence of ageing on the material fracture strength, kinetic models previously applied in this paper have been analyzed considering K_Q instead of the DP. These expressions have allowed to relate the evolution of the K_Q over time as a function of the temperature. As can be observed (Fig. 12), the model proposed by Carrascal et al. (Carrascal et al. 2018) provides the highest coefficient of determination R2 after the fitting of the free parameters when K_Q in MD is considered. However, when K_Q in CD was studied the highest coefficient of determination R2 was obtained by the model of Emsley et al. (Emsley et al. 1997).

The correlation between the DP and K_Q was also obtained, see Fig. 13. As can be seen, there is a linear relation between the DP and K_Q which depends on the fiber angle (MD and CD).

The correlations between DP and K_Q for the orientations CD and MD are shown in Table 5. The coefficient of determination R2 is higher than 0.9 in both cases. This result is an evidence in favor of the use of K_Q to quantify the loss of mechanical strength of insulation paper in power transformers during thermal stress.

Table 5. Stress intensity factor vs DP of Kraft paper aged at 130^{o}C

	Mathematical correlation	R^2
Cross direction (CD)	$DP = 263.93*K_Q + 30.52$	0.9656
Machine direction (MD)	$DP = 93.22*K_Q + 143.84$	0.9152

Conclusions

The DP of the Kraft paper studied in this article has demonstrated to be an efficient tool to measure the loss of mechanical strength suffered by cellulosic materials over time as a function of the temperature. However, due to some practical difficulties with its determination for some new papers after their drying, as well as the increasing interest in synthetic dielectric materials replacing cellulosic insulation, it is needed the establishment of alternative methods to DP to quantify the deterioration of insulation solids accurately. This role can be played by the properties obtained from tensile and fracture toughness tests as it has been demonstrated in this work.

Double-edged notched specimens were tested in tension to study their fracture toughness. The specimens analyzed did not fulfil the requirements to consider K_Q as a geometry-independent measure of fracture toughness, consequently K_Q is not equal to K_{Ic} but it is a valid characterization of the material fracture toughness for the thickness of the specimen.

The stress intensity factor (K_Q) allows to quantify the influence of thermal ageing on the material fracture strength. It was obtained that the K_Q was reduced below 32% of its initial value in the first 200 hours which was in accordance with the fractographic study. The fractographies showed how the fibres' strain in the rupture section is considerably reduced during thermal ageing.

Additionally, kinetic models were applied considering the K_Q which made possible to relate the evolution of the K_Q over time as a function of the temperature.

Finally, the linear correlation between the DP and K_Q, which depends on the fiber angle (MD and CD), was also obtained.

Declarations

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Figures

Figure 1

Vessel for thermal ageing

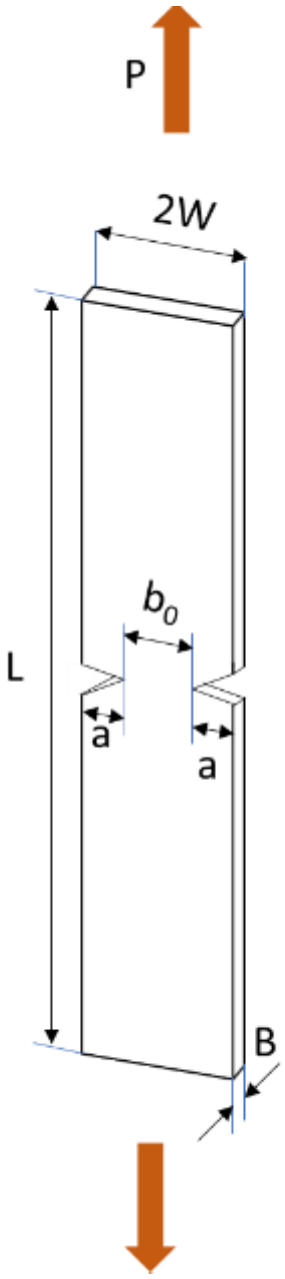


Figure 2

Deep, double-edged, notched specimen tested in tension for experiments

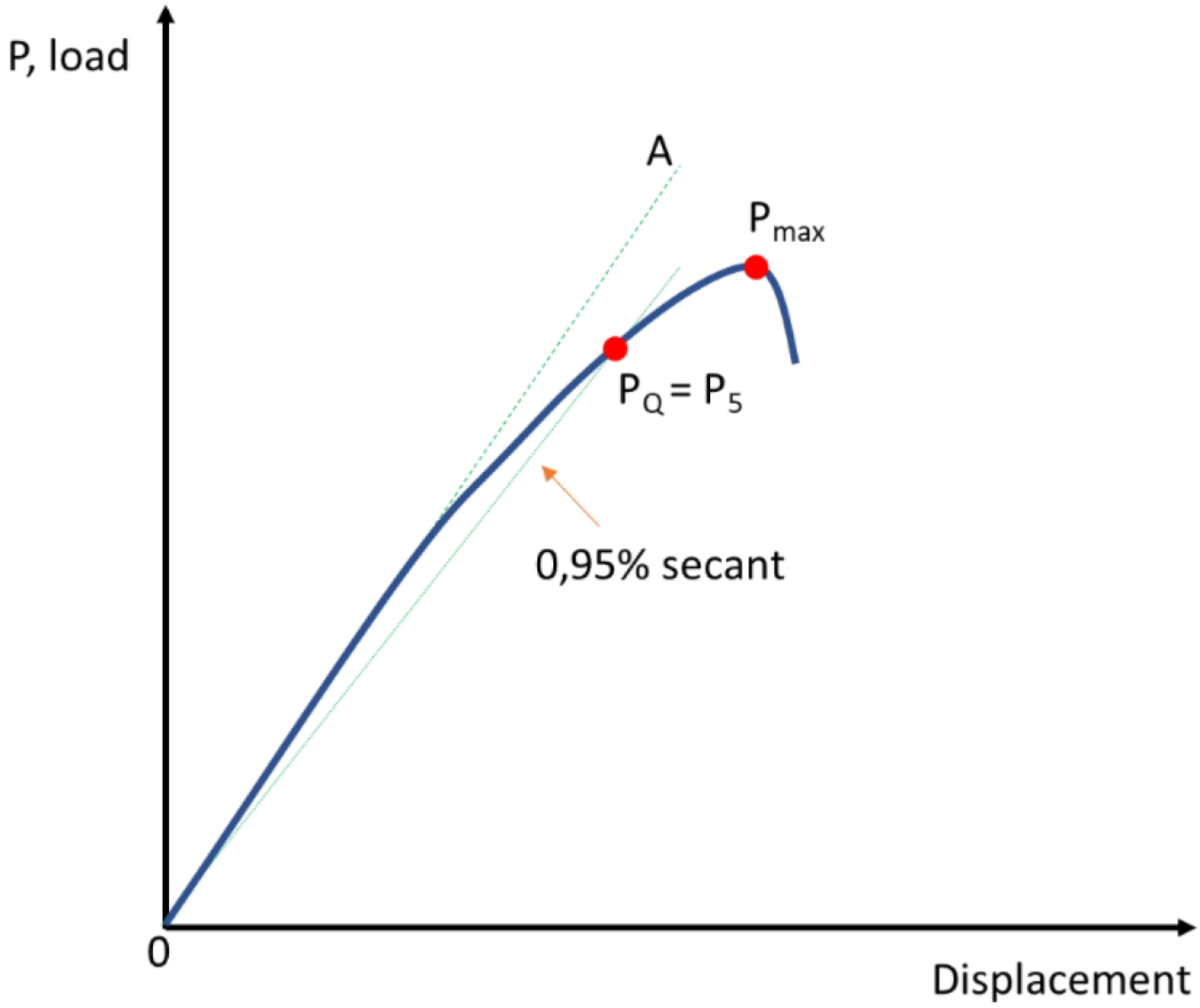


Figure 3

Load-displacement curve mode I

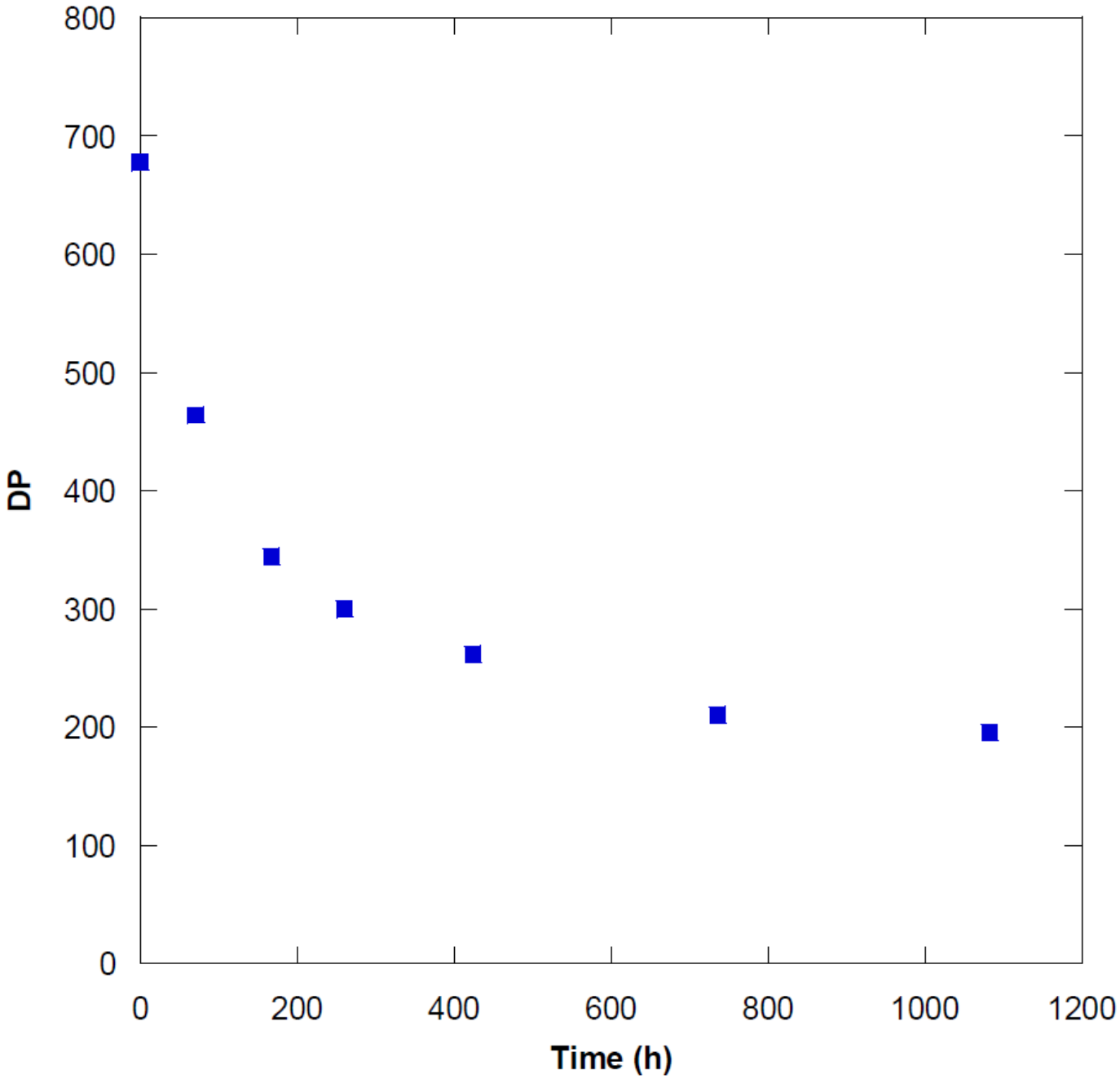
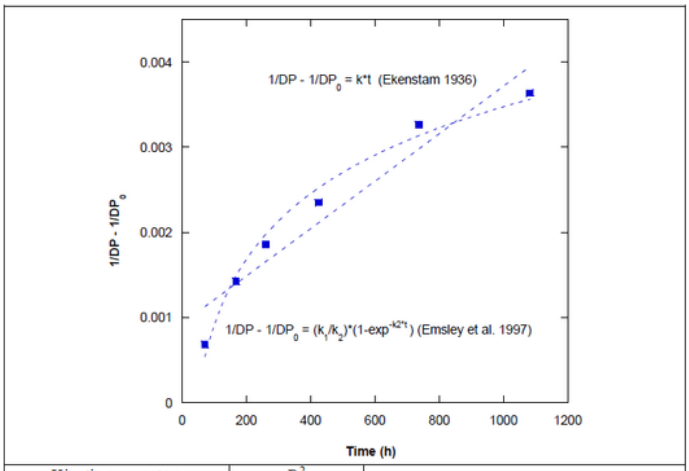
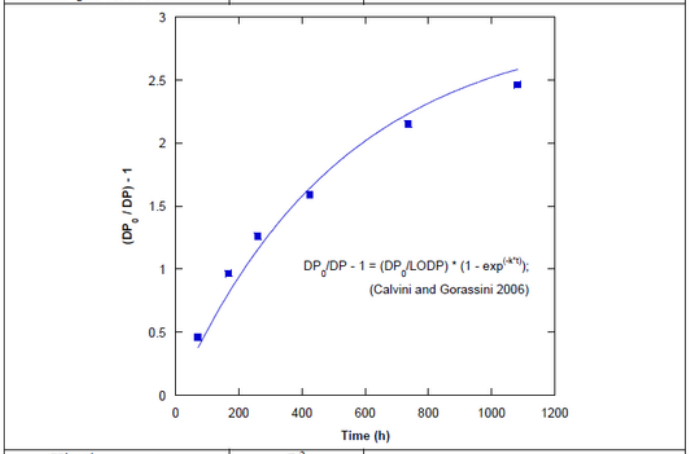


Figure 4

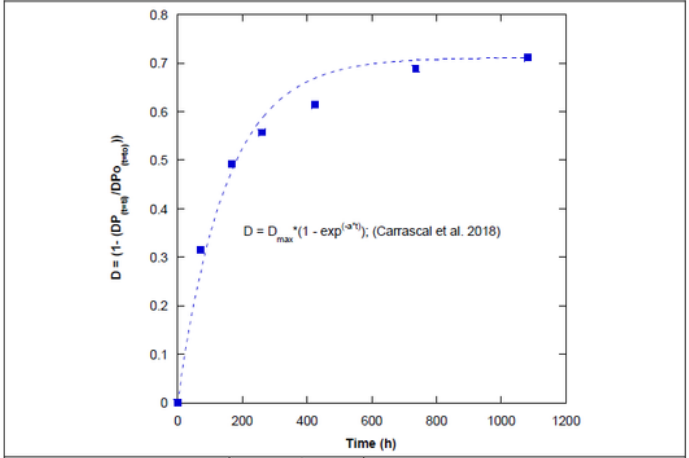
Evolution of the DP as a function of time at 130oC



Kinetic parameters	R^2	
$k = 2.79*10^{-6}$	0.9230	(Ekenstam 1936)
$k_{10} = 9.54*10^{-6}$ $k_2 = 2.46*10^{-3}$	0.9957	(Emsley et al. 1997)



Kinetic parameters	R^2	
$k = 1.49*10^{-3}$	0.9678	(Calvini and Gorassini 2006)



Kinetic parameters	R^2	
$a = 6.65*10^{-3}$	0.9832	(Carrascal et al. 2018)

Figure 5

Kinetic models based on the DP

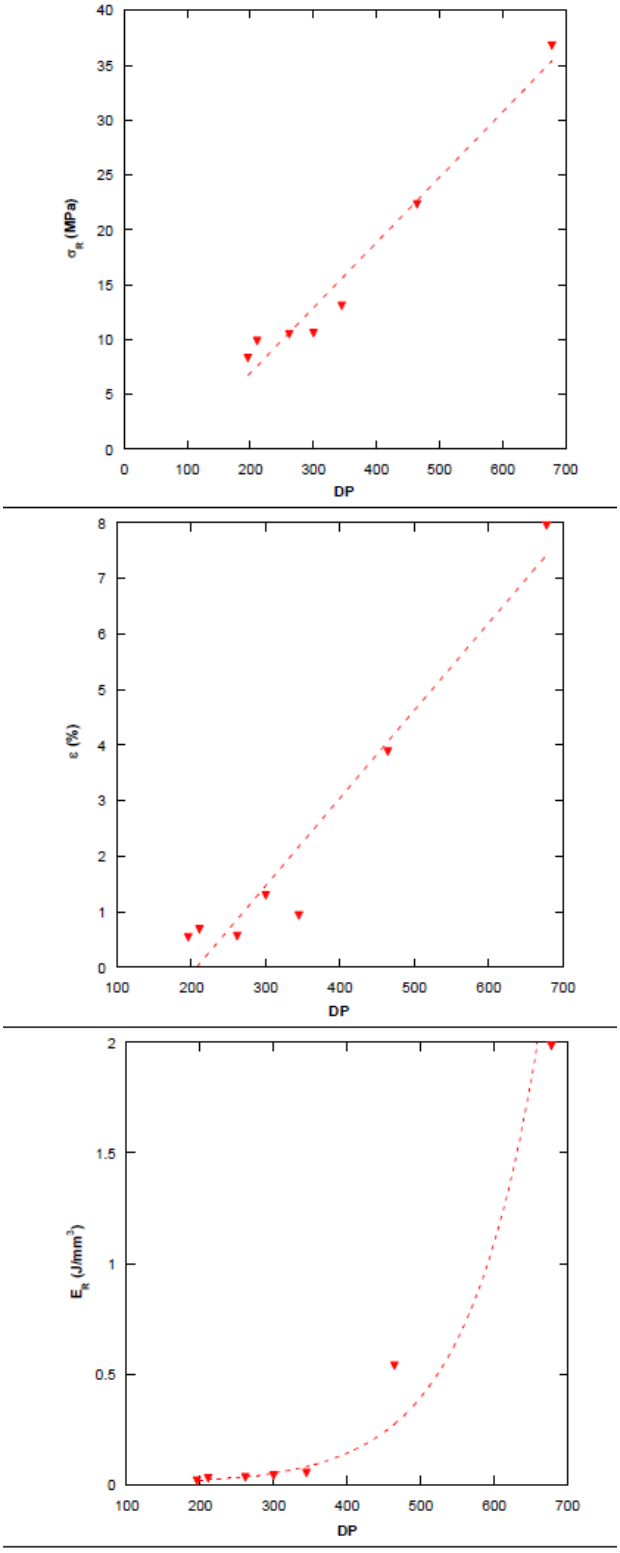


Figure 6

Mechanical properties vs DP in CD

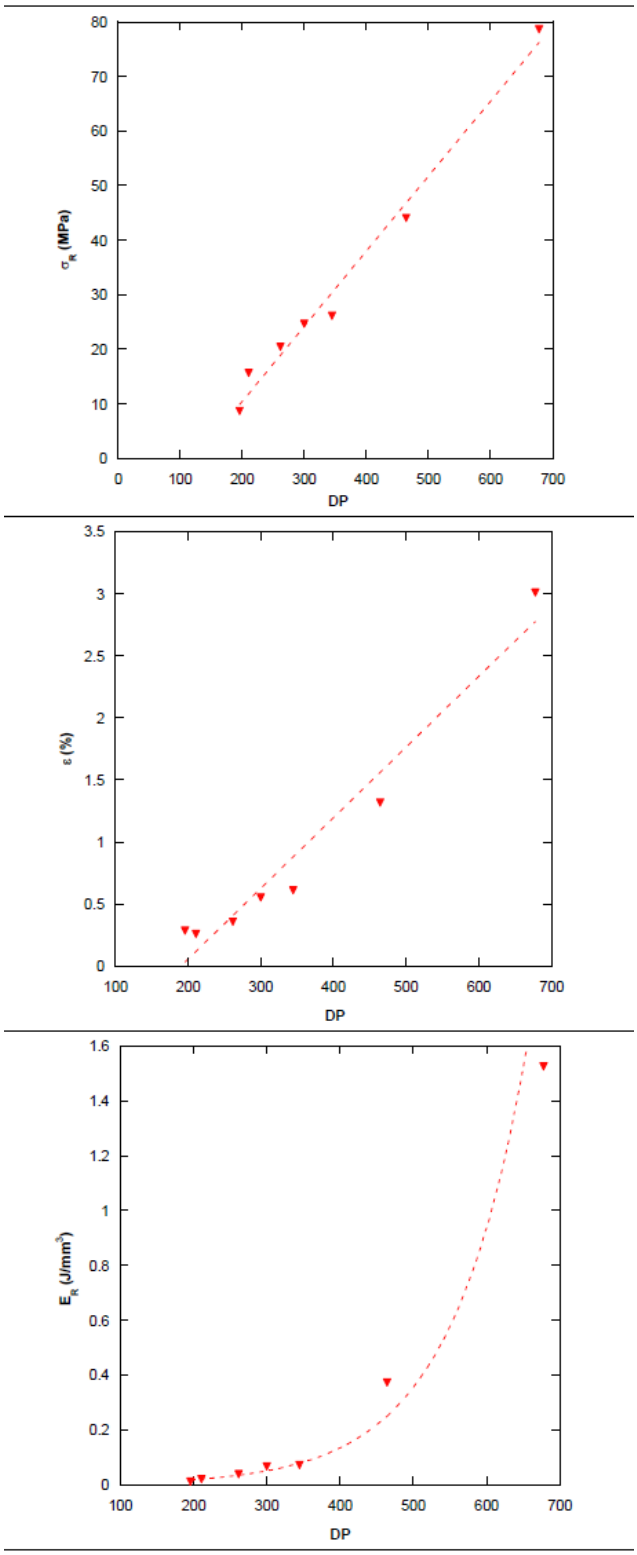


Figure 7

Mechanical properties vs DP in MD

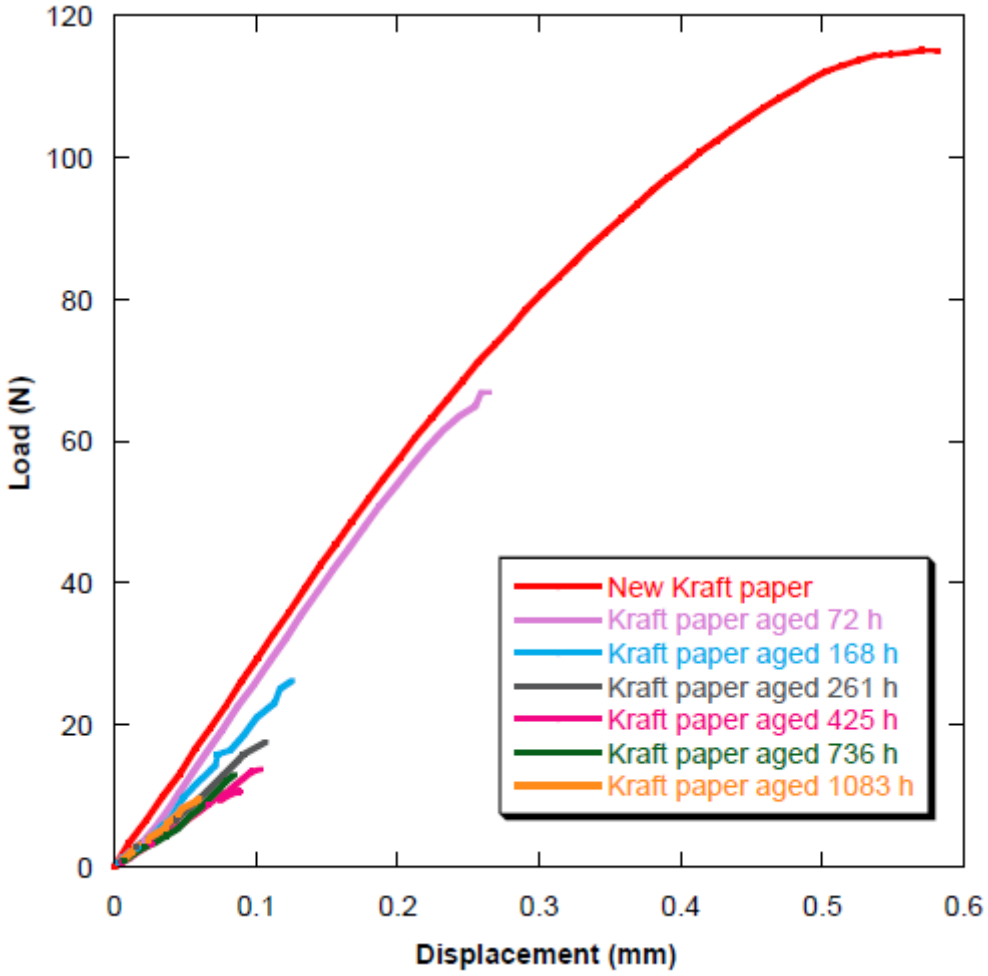


Figure 8

Influence of the t of ageing on load and displacement in MD

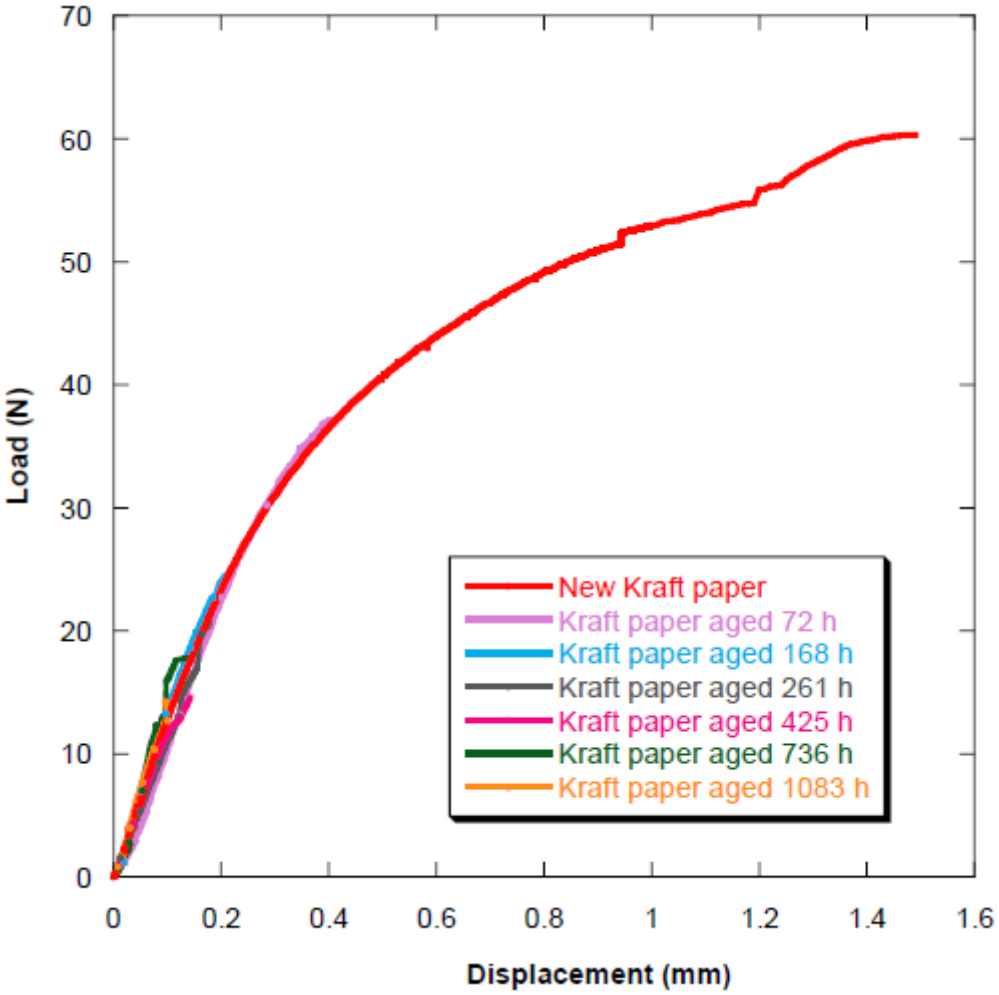


Figure 9

Influence of the t of ageing on load and displacement in CD

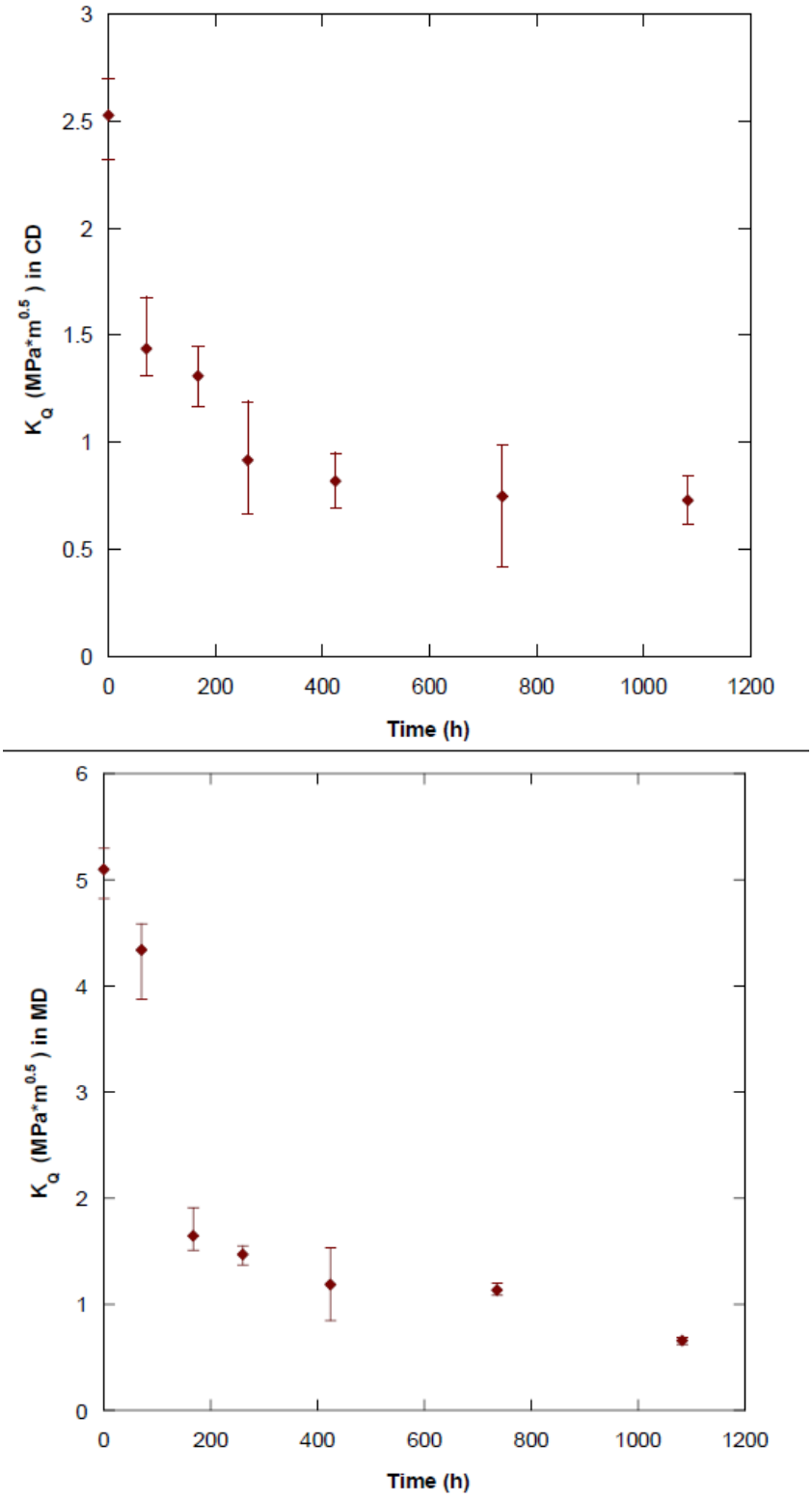


Figure 10

KQ in MD and CD as a function of the ageing t

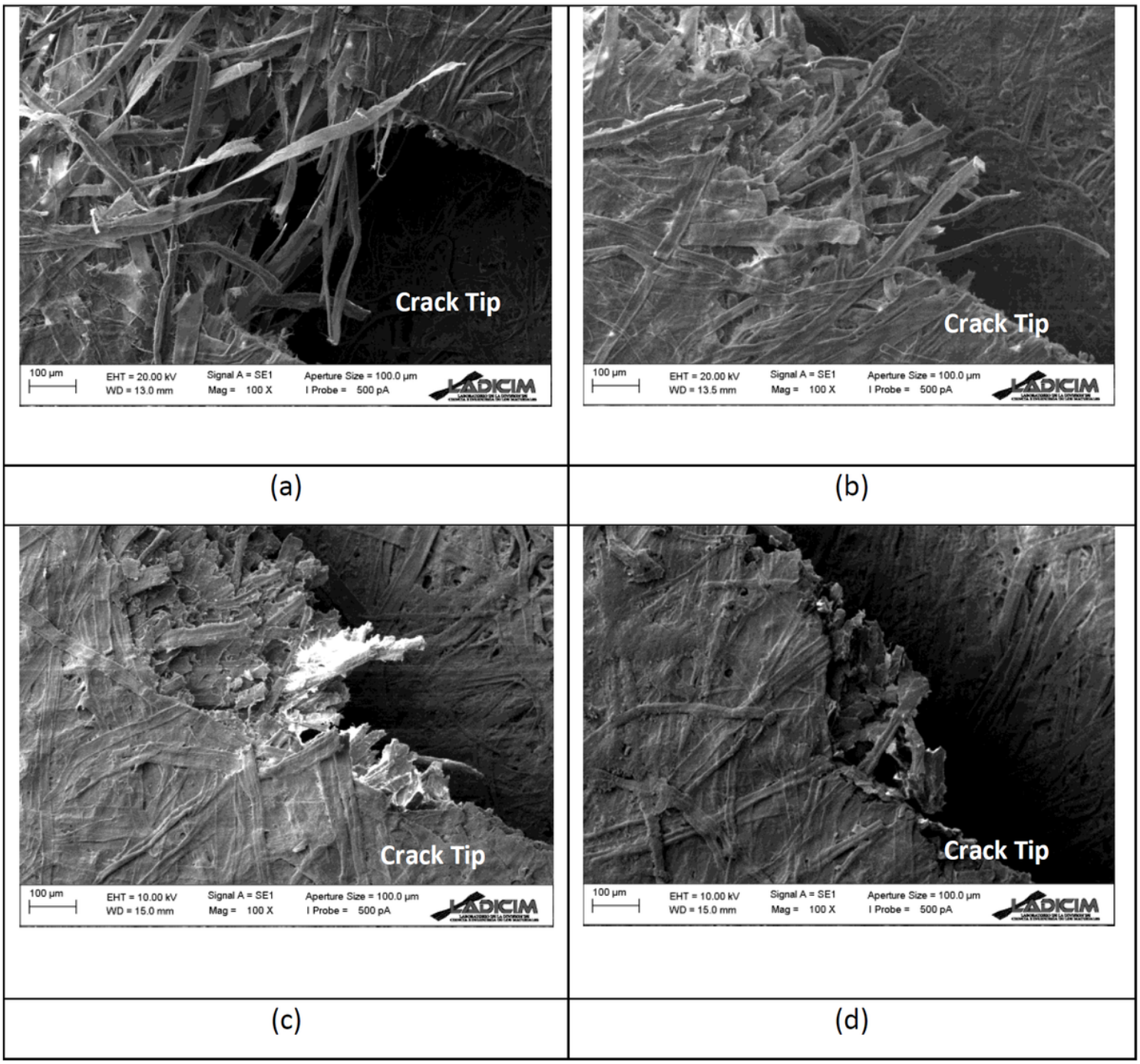
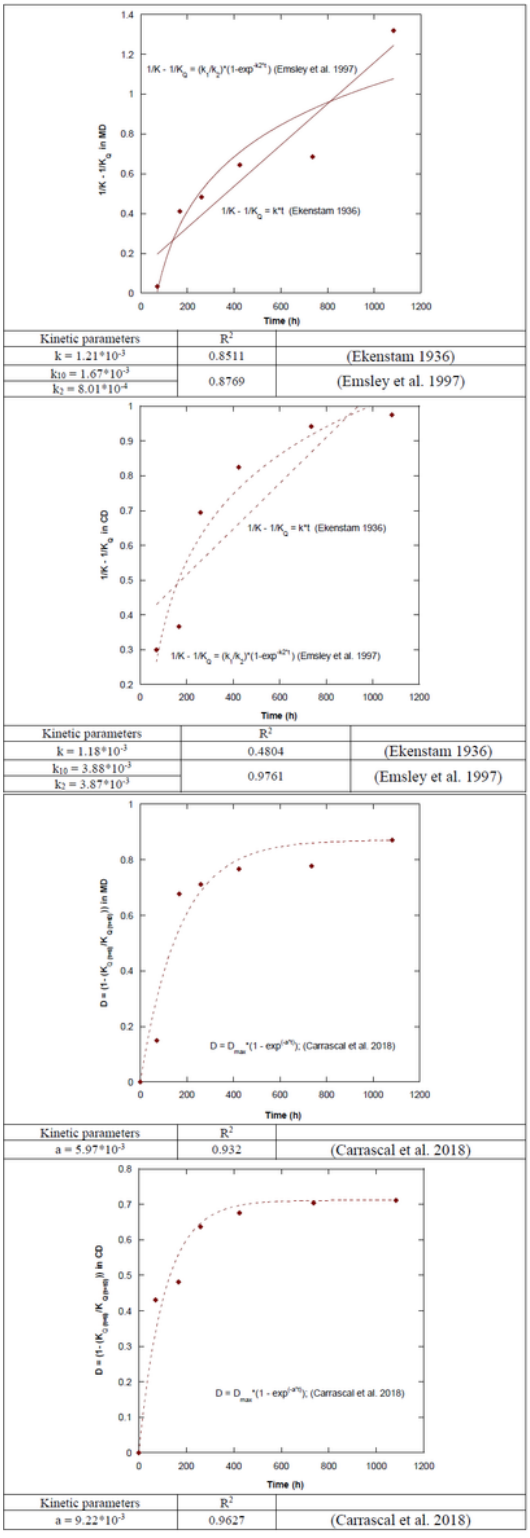


Figure 11

a New Kraft paper (KQ in MD = 5.1 MPa*m0.5), b Kraft paper aged during 72 h at 130ºC (KQ in MD = 4.34 MPa*m0.5), c Kraft paper aged during 168 h at 130ºC (KQ in MD = 1.65 MPa*m0.5), d Kraft paper aged during 1083 h at 130ºC (KQ in MD = 0.66 MPa*m0.5)



Kinetic parameters	R^2	
$k = 1.21*10^{-3}$	0.8511	(Ekenstam 1936)
$k_{10} = 1.67*10^{-3}$ $k_2 = 8.01*10^{-4}$	0.8769	(Emsley et al. 1997)

Kinetic parameters	R^2	
$k = 1.18*10^{-3}$	0.4804	(Ekenstam 1936)
$k_{10} = 3.88*10^{-3}$ $k_2 = 3.87*10^{-3}$	0.9761	(Emsley et al. 1997)

Kinetic parameters	R^2	
$a = 5.97*10^{-3}$	0.932	(Carrascal et al. 2018)

Kinetic parameters	R^2	
$a = 9.22*10^{-3}$	0.9627	(Carrascal et al. 2018)

Figure 12

Kinetic models based on the KQ

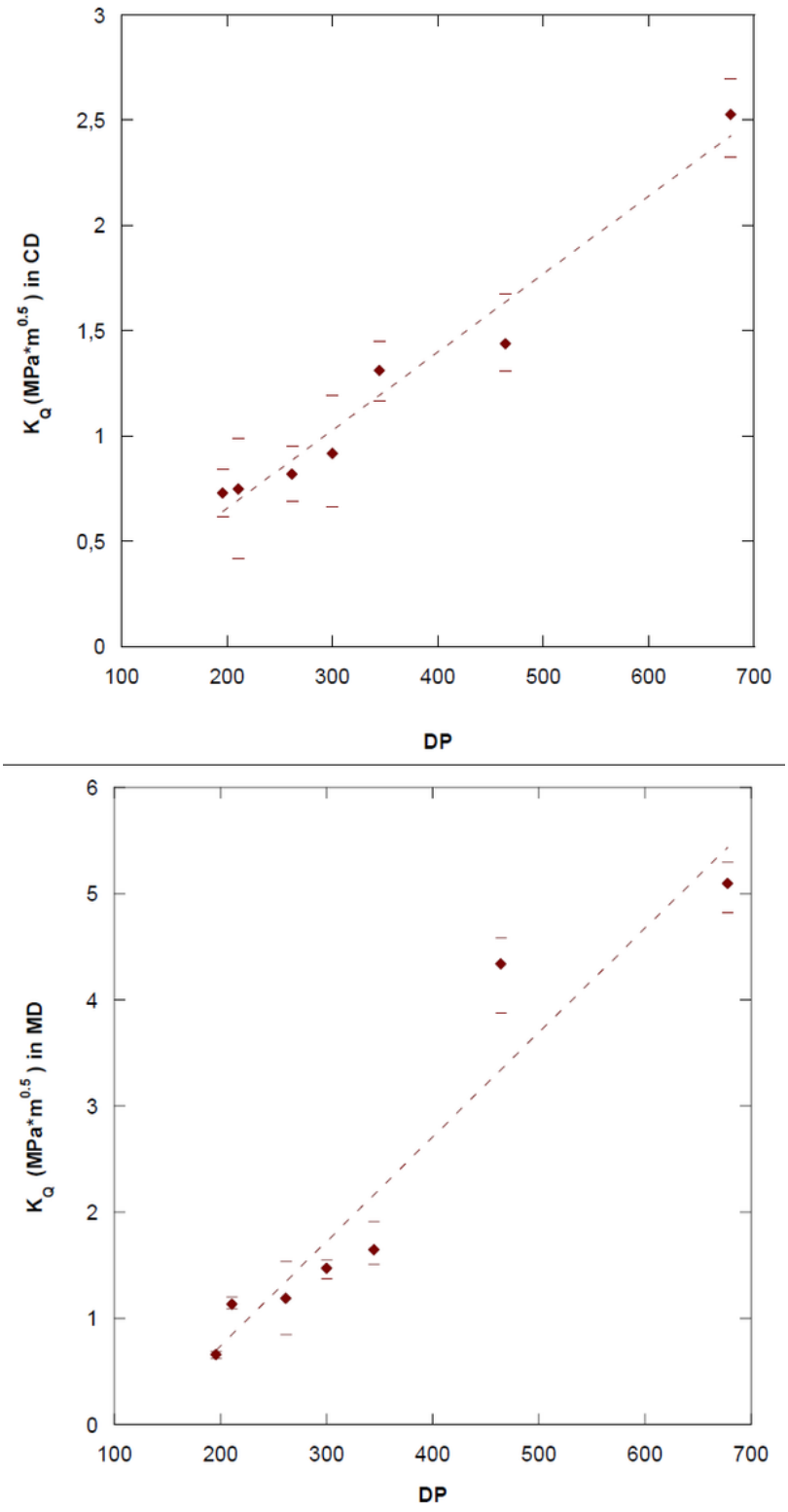


Figure 13

KQ vs DP in MD and CD

Supplementary Files

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- GRAPHICALABSTRACT.png