# Investigation of the degradation of a wood pulp-cotton presspaper in different biodegradable oils


Cristina Méndez Gutiérrez
*Electrical and Energy Engineering Department*
*University of Cantabria*
Santander, Spain
cristina.mendez@unican.es

Jaime Sanz Asensio
*Electrical and Energy Engineering Department*
*University of Cantabria*
Santander, Spain
uc26239@alumnos.unican.es

Cristian Olmo Salas
*Electrical and Energy Engineering Department*
*University of Cantabria*
Santander, Spain
cristian.olmo@unican.es

Fernando Delgado
*Electrical and Energy Eng. Department*
*University of Cantabria*
Santander, Spain
delgadof@unican.es

Pedro Quintanilla Cavia
*Electrical and Energy Eng. Department*
*University of Cantabria*
Santander, Spain
quintanillap@unican.es

Riccardo Maina
*Sea Marconi Technologies*
Torino, Italy
riccardo.maina@seamarconi.it

Alfredo Ortiz Fernández
*Electrical and Energy Eng. Department*
*University of Cantabria*
Santander, Spain
ortizfa@unican.es



*Abstract*— **New insulating materials, as biodegradable oils and upgraded paper, need to be investigated. In this paper, the ageing of a wood pulp – cotton presspaper is studied. Solid insulation is impregnated and aged with three different fluids: a mineral oil and two vegetable oils, from sunflower and soybean. Both oils and paper were dried before the ageing process, which was carried out at 150ºC for 732 hours in iron vessels. Degradation of oils is analysed through the measurement of their breakdown voltage, dielectric dissipation factor (tan δ), resistivity, moisture content and acidity. Deterioration of presspaper is quantified by its polymerization degree (DP), moisture content and dielectric dissipation factor. Results showed that the dielectric properties of oils are negatively affected by the ageing, since the breakdown voltage and resistivity were reduced, whereas the dissipation factor increased. Also, acidity increased, especially in the vegetable oils. In the case of the solid insulation, its tanδ increased with the ageing, despite the reduction of its moisture content. DP was reduced, reaching the end-of-life criteria (DP<200) in the ageing with mineral oil (134) whereas it remained at higher values for the sunflower (206) and soybean (216) oils.**



**Keywords—power transformers, natural esters, presspaper, thermal ageing**


## I. Introduction

Electricity demand is constantly increasing. Changes in the electrical system, including renewable energies and distributed generation, make it necessary to update the transmission and distribution lines. Power transformers are essential to ensure an efficient energy transport and safe consumption, so it is important to guarantee their correct operation while we improve their technology and make them more sustainable. Despite their high efficiency, these machines, as all the electrical equipment, have losses during their operation that cause an increase of the temperature. Very high temperatures can damage the transformer' components, so it is common to use cooling fluids, especially in the big machines. Traditionally, mineral oil has been used for cooling and, also, dielectric isolation of the transformers. Until these days, mineral oil was cheap, and its properties were accurate for their use in electrical equipment. Nevertheless, with the new power and environmental requirements, mineral oil is no longer suitable, due to its low biodegradability and low flash and fire points. Natural and synthetic esters are being considered to replace traditional oil. These new fluids are biodegradable and have high flash and fire points, while having good dielectric capacity. The main challenge for the use of these oils is their different nature, that makes that their physico-chemical properties notably differ from that of the mineral oil [1].

It is well known that these fluids have good dielectric capacity, high flash and fire points and that their main throwback is their high viscosity [2], which affects their cooling capacity. Nevertheless, this problem can be solved with a proper design of the transformer [3].

In view of the suitable properties of the esters for their use in power transformers, research works in the last years have been focused on understanding their degradation after several years of exposure to thermal, mechanical, and electrical stresses. These works study not only the evolution of the oil, but also the deterioration of the most critical component of the transformer, that it is the cellulose used as solid insulation. The ageing of the insulation system is affected by different factors, such as the temperature, oxygen, electrical failures or moisture content [4]. Different studies have been carried out with both natural and synthetic esters, some of them comparing these alternative oils [5]. Most of the works are focused on the ageing of kraft or thermally upgraded kraft (TUK) papers. They have found that kraft paper degradation is lower when ageing with the esters than with the mineral oil [6]. This better behaviour of the esters is explained by their hygroscopic capacity [7] and by the type of acids generated during their aging [8]. Esters usually have higher moisture content than mineral oil and they tend to absorb water. This characteristic, far from being a disadvantage when subjected to high temperatures, make them capable to absorb water from the cellulose through the hydrolysis process, which reduces the cellulose degradation. Also, oils increase their acidity during their ageing. Acids generate by the mineral oil are low molecular, whereas esters generate high molecular acids. Different works noticed that the high molecular acids do not damage the cellulose as the low molecular acids do.

It has been found that there are few references studying solid insulation different from kraft or TUK paper. Also, most of the works are focused on the comparison between mineral and one natural or synthetic ester, but there are hardly any references with different vegetable oils. Due to the differences between the materials and ageing tests carried out by different authors, it is difficult to draw conclusions about which


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vegetable oil is more suitable for being used with a certain solid insulation. Because of that, in this work we study the ageing of a presspaper mixture of wood pulp and cotton, especially suitable for uneven surfaces due to its flexibility. This presspaper was impregnated and aged with three different oils. One mineral oil and two natural esters, from soybean and sunflower. Degradation of both oils and paper were considered, through different parameters, including dielectric and physico-chemical properties.

## II. Experimental Procedure

### A. *Materials and accelerated thermal ageing*

A mixture of wood pulp and cotton presspaper was used as solid insulation. Its properties are shown in Table I. Presspaper was cut in strips and squares of sizes suitable to carry out the desired measurements.

Three commercial dielectric oils were considered in this work. Two natural esters, derived from soybean and sunflower, and a mineral oil, whose properties are listed in Table II.

Both oils and paper were dried before the impregnation and ageing process. Oils were dried for 24 hours at 60ºC in a vacuum oven in cycles of 4 hours of vacuum and 1 hour at 500 mbar in a nitrogen atmosphere. Presspaper was dried 3 hours at 105ºC in an air circulating oven.

Then, both materials and copper were introduced in an iron vessel, with a 1 g paper / 0.5 g copper / 10 g oil ratio. After filling the vessels, they were sealed with nitrogen and placed in an oil-circulating oven at 60ºC for 1 hour to carry out the impregnation of the presspaper with each oil.

Finally, the temperature was increased up to 150ºC and ageing process started. Samples were analysed after different ageing periods: 15, 48, 72, 168, 228 and 732 hours, when the paper of one of the vessels reached its end of life (DP < 200).

TABLE I. Properties of the presspaper

| Property | Specification |
|---|---|
| Density [g/cm$^3$] | 1 |
| Tensile strength (machine direction) [MPa] | 91 |
| Elongation (machine direction) [%] | 2,8 |
| Moisture content [%] | < 8 |
| Electric strength in air [kV/mm] | 7 |
| Electric strength in oil [kV/mm] | 55 |

TABLE II. Properties of the oils

| Property | Mineral | Soybean | Sunflower |
|---|---|---|---|
| Density 20ºC [g/cm3] | 0.842 | - | 0.91 |
| Viscosity. 40ºC [cSt] | 10.3 | ≤ 50 | 39.2 |
| Flash point [ºC] | 170 | > 250 | 330 |
| Pour point [ºC] | - 48 | ≤ -10 | - 25 |
| Acidity [mg KOH/g] | < 0.01 | ≤ 0.6 | 0.05 |
| Water content [mg/kg] | 15 | ≤ 200 | 150 |
| Dissipation factor 90 °C | 0.00074 | ≤ 0.05 | 0.03 |
| Dielectric Breakdown [kV] | 35 | ≥ 35 | 65 |

### B. *Oil tests*

Different tests were carried out in the oils to analyse their properties and degradation, including dielectric and physico-chemical parameters.

Regarding dielectric properties, AC breakdown voltage (BDV) was measured following the IEC 60156 standard. BDV was determined by a bA100 dielectric tester, in which the test vessel with semispherical electrodes separated 2.5 mm was filled with 400 ml of oil. Each sample is subjected to six breakdown tests with 2 minutes of magnetic stirring between each measurement. A BAUR DTL 2a was used to determine the dielectric loss factor (tan δ), permittivity (ε), and resistivity of each oil. All the parameters are measured in the same test at 90ºC, according to the IEC 60247 methodology.

Due to effect of moisture content on the dielectric properties and, also, in the ageing of solid insulation, this parameter was controlled by Karl-Fischer titration, following the IEC 60814 standard. Also, the acidity was measured since it is considered as one of the most feasible indicators of oil degradation. Acidity was determined by automatic potentiometric titration according to the IEC 62021 methodology.

### C. *Presspaper tests*

Different properties of the presspaper were measured in order to analyse its degradation and the effect it has on the dielectric response.

The dielectric analysis is mainly focused on the study of the dielectric dissipation factor (tan δ). It was measured using a combination of the Frequency Domain Spectroscopy (FDS) and Polarization/Depolarization Current (PDC) techniques. Tests were carried out with the analyzer Spectano 100 in combination with the sample holder DSH100, at room temperature (25ºC) for a with a frequency range from 17 mHz to 5 kHz.

Also, the degree of polymerisation (DP) of the cellulose was determined following the ASTM D4243 standard. This parameter is one of the most commonly used to determine the cellulose degradation, considering a DP lower than 200 as the end of life of solid insulation.

## III. Results

### A. *Acidity and moisture content*

Acidity results, presented in Fig.1, showed that it increased with the ageing, following a potential relationship in all the oils. The acidity of soybean oil was the highest one, despite its initial value was similar to that of sunflower oil. As expected, the mineral oil had the lowest acidity.

The moisture content of both the oils and presspaper is shown in Fig.2. As it was expected, the natural esters had a higher moisture content than the mineral oil. Also, at the beginning of the ageing process the moisture in the natural esters increased, whereas it remained almost constant in the mineral oil. According to previous studies, the increase of water in the natural esters is due to the hydrolysis process [7] that does not occur in the mineral oil. If considering the cellulose, it is well known that it produces water with its degradation. In the case of the ageing with natural esters, this water was partially absorbed by the oils, which caused that at the end of the experiment, the moisture in cellulose was reduced. Conversely, it increased with the mineral oil.

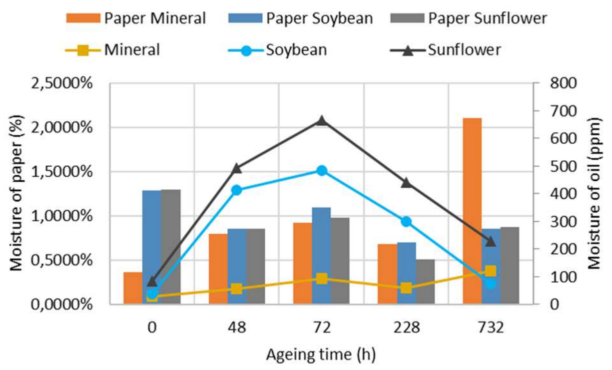


Fig 1. Moisture content of oils and presspaper.

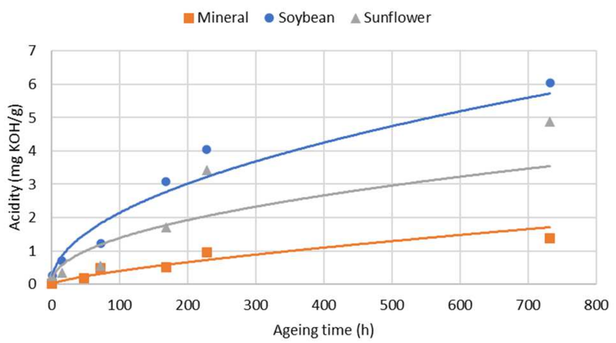


Fig 2. Acidity of oils.

## *B. Dielectric properties*

BDV showed that the dielectric capacity of all the oils was reduced with the thermal ageing. This reduction was more pronounced in the mineral oil (78.1%) than in the soybean (55.5%) and sunflower (52.2%) oils. Also, it is interesting to highlight that the two natural esters had similar dielectric capacity and their BDV was always higher than that of the mineral oil.

Also, the tan δ results showed a decrease in the electrical insulation capacity of the oils with the degradation, since the tan δ increases with the ageing (Fig.3). As expected, tan δ of the natural esters was notably higher than that of the mineral oil. Resistivity showed the same tendency than the other electrical parameters, being reduced by the ageing and higher in the mineral oil. Permittivity of oils remained almost constant. The degradation caused a small increased of this parameter only at the end of the ageing of the natural esters (3.6%). As expected, permittivity of the mineral oil (2.1) was lower than that of the natural esters (2.8).

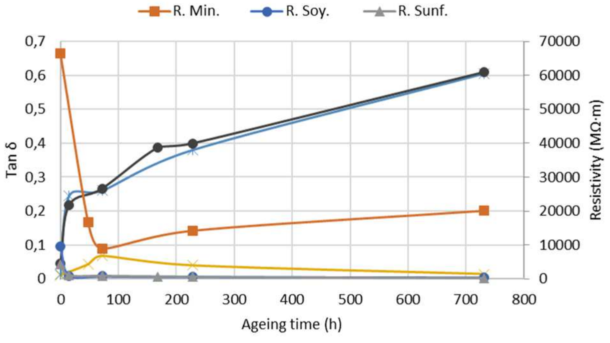


Fig 3. Dielectric los factor and resistivity of oils.

Tan δ of the impregnated presspaper increased with the ageing for all the combinations studied, as presented in Fig. 4 and Fig. 5. In the case of the mineral oil, this increase was more relevant at the beginning of the ageing. Once the presspaper was highly degraded, its tan δ remained almost constant. Conversely, in the tests carried out with the natural esters, the tan δ of the presspaper increased during all the ageing. Also, it is interesting to analyse the different shapes of the tan δ response depending on the oil that impregnated the paper. Natural esters showed higher tan δ at low frequencies and this parameter decreased significantly with the increase of the frequency. Mineral oil also showed higher tan δ at low frequencies, but the difference between the tan δ at the lowest and highest frequencies was not so pronounced. This could be due to the behaviour of the mineral oil itself: it had lower moisture content and it showed smaller changes on its tan δ response with the ageing in comparison with the natural esters. This behaviour agreed with the conclusions obtained when comparing the response of the presspapers impregnated with different fluids. As shown in Fig.6, the impregnating fluid had a great influence on the dielectric response, since the lower tan δ of the mineral oil led to lower tan δ of the presspaper impregnated with this fluid.

## *C. Polymerisation degree*

Considering the differences in the chemical structure of the oils analysed, which affect their dielectric and chemical properties, it is difficult to draw conclusions about the degradation of the presspaper from the results previously shown. Thus, the polymerisation degree of the cellulose was measured, and the results are presented in Fig. 7. Prior to ageing, the DP of the presspaper was 993. This value is slightly lower than the expected (1200), probably due to a little degradation caused by the drying process and, also, because the constants used to calculate the DP value are more accurate for Kraft paper. DP was reduced with the degradation of the cellulose and reached a value of 134 for the last point of the mineral oil. The presspaper had higher DP when aged with the natural esters than with the mineral oil along the ageing process. At the beginning of the ageing differences between oils were less significant, but they increased at the end of the process. Despite the ageing with both natural esters were similar and better than with mineral oil, sunflower seemed to protect a little bit more the cellulose (DP = 229) than the soybean oil (DP = 216), which agree with the highest acidity showed by the soybean oil.

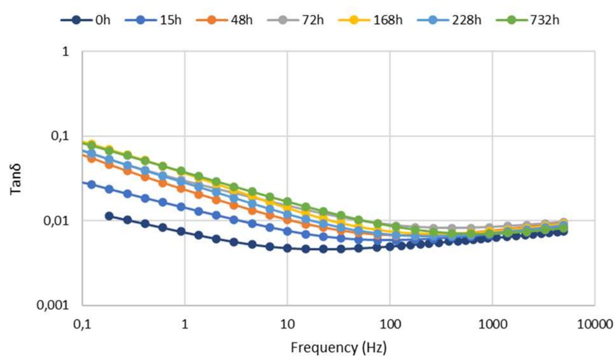


Fig 4. Dielectric loss factor of the presspaper aged with mineral oil.

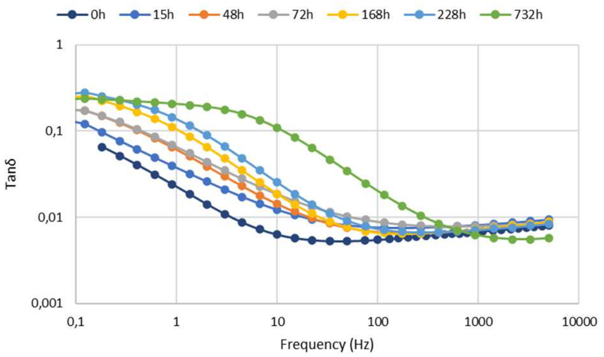


Fig 5. Dielectric loss factor of the presspaper aged with soybean oil.

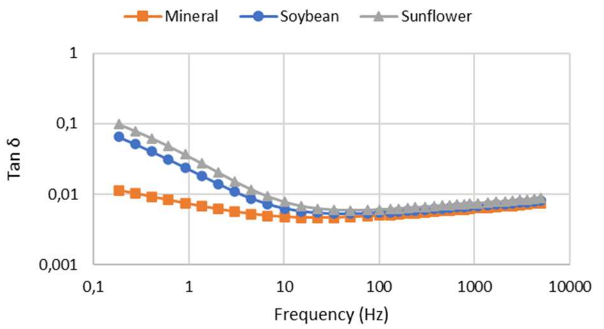


Fig 6. Dielectric loss factor of the presspaper with different oils.

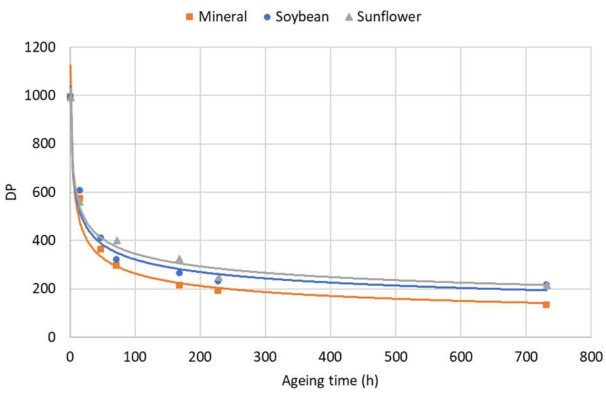


Fig 7. Polymertisation degree of the presspaper aged with different oils.

## IV. Conclusions

Ageing of a wood pulp – cotton presspaper was analysed in the presence of a mineral, sunflower, and soybean oils. Degradation of the oils and the cellulose was studied through the measurement of dielectric (BDV, tan δ, ε and resistivity) and physico-chemical (acidity, moisture, and DP) properties.

The acidity of all the oils increased with their degradation, being always higher in the natural esters. At the end, soybean had the highest acidity, which suggested a higher degradation. The moisture content of the mineral oil remained almost constant, whereas it increased in the esters. The moisture of the presspaper immersed in mineral oil increased, whereas it decreased when immersed in vegetable oils.

The oils' dielectric capacity was reduced with the degradation. BDV decreased in all the oils, but especially in the mineral oil, whereas this reduction was similar in the natural esters. Tan δ increased and resistivity decreased, and the mineral oil showed better results, due to the absence of polar compounds. Tan δ of the paper increased with the ageing and it was lower when impregnating with mineral oil.

The degradation of the cellulose could not be inferred from the results of the oils, due to their different nature. According to the DP results, the presspaper suffered the highest degradation with the mineral oil and the lowest with the sunflower. The difference between sunflower and soybean oil was not significant, but agreed with the acidity results.

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