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Analysing the Suitability of Using Different Biodegradable Fluids for Power Transformers with Thermally Upgraded Paper

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Abstract: Mineral oil has been used for many years in various electrical equipment, including transformers, as a cooling and insulation medium. However, its low biodegradability and poor performance in terms of fire protection have prompted the search for fluids to replace it, with vegetable oils being prominently considered. In this study, the dielectric, chemical, and physical properties of four vegetable oils obtained from different seeds (sunflower, rapeseed, soybean, and palm) and a biodegradable synthetic fluid are analysed throughout their lifespan in transformers. Their performances are compared with a traditional mineral oil to assess which one is more suitable for use in transformers employing this type of paper. To achieve this, the fluids were subjected to thermal ageing in combination with copper and a thermally upgraded kraft (TUK) paper, with its degradation controlled by measuring the degree of polymerisation. The results demonstrate that the origin of the vegetable oils affects their properties and degradation rates. It was found that most of the alternative fluids are suitable for use in transformers with the TUK paper and that they can increase their lifespan.

Keywords: biodegradable fluids; esters; dielectric insulation; degradation; power transformers

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1. Introduction

In recent years, protecting the environment has become increasingly important as a foundation for sustainable development. The energy sector, one of the largest CO_2 emitters [1], plays a key role in sustainable development goals [2]. Therefore, different ways to reduce the environmental impact of this sector are being studied, from generation using renewable energy sources and more efficient storage systems [3,4] to consumer contributions [5,6]. It is also essential to improve the transportation and distribution of electricity [7], in which substations play a fundamental role.

Additionally, the European electrical system is ageing, with about a third of its networks being over 40 years old [8]. Given this context and the existing need for renovation, it is an opportunity to adapt it to the environmental requirements without incurring greater costs [9].

Within electrical substations, transformers are the primary components. These require electrical insulation and a cooling system, usually composed of thousands of litres of mineral oil [10] and solid cellulose-based insulation [11]. Sustainable alternatives to this kind of oil have been sought for years, with natural and synthetic esters being prominent [12,13]. Introducing these biodegradable fluids, derived from renewable resources such as crops, in place of traditional petroleum-derived oils would represent a significant advancement in the sustainability of the electrical system. Moreover, it would help reduce the risk of fires [14] and contamination in the event of spills, thus contributing to environmental protection.

As a result, numerous authors have researched these fluids, focusing on their long-term behaviour and the degradation of solid insulation, as these conditions determine the transformer's lifespan [15]. Given the high cost of this equipment and its importance in the electrical system, understanding material ageing is crucial for proper maintenance and ensuring greater durability [16]. Furthermore, if replacing the cooling oil compromises the equipment's durability, any sustainability enhancements would be nullified, as it would lead to the need for transformer replacements sooner and more frequently than expected.

These studies consist of laboratory analysis of the fluid and paper property evolution under thermal stresses over time, as it happens in the actual transformers, focusing on those with an impact on the insulating and cooling capacities and the integrity of the fluids and papers. However, to date, studies have typically focused on one or two esters combined primarily with conventional paper, with significant variations in test conditions between studies. This makes it difficult to compare results and determine the best alternative. Indeed, upon analysing the results obtained in different studies, it is observed that they vary greatly and are even contradictory. For instance, concerning the dielectric properties of the fluids, while one study found that the breakdown voltage (BDV) did not change with ageing [17], another study conducted with the same fluid and the same ageing temperature applied for a shorter period found a decrease in the BDV of 35% [18]. The same is true for the dielectric dissipation factor (tan δ), with values found in references varying widely from $6.5{\cdot}10^{-2}$ [19], $4.5{\cdot}10^{-3}$ [18], to $3.7{\cdot}10^{-4}$ [20], for the same fluid. Regarding chemical properties, although an increase in fluid acidity with ageing is observed in all studies, this increase differs from one study to another. For example, in [21] and [22], acidity increased by 5000%, but the ageing times were 7896 and 3186 h, respectively. These differences also apply to paper degradation; in two ageing studies at 120 °C, with the same paper and ester, a reduction of 70% in the polymerisation degree (DP) was found after 2000 h in one study [23], while in the other [17], the reduction was 60% after 8600 h.

It is essential to analyse the main alternatives under the same conditions so that results are comparable. Additionally, it is important to study the combination of these fluids with new cellulose materials, which have greater thermal resistance [24,25] and allow for better equipment performance.

This study investigates the five main commercial esters along with widely used thermally upgraded kraft (TUK) paper under thermal stress conditions. The main properties of the fluids and paper degradation are analysed to determine the best combination of materials.

2. Materials and Methods

In this section, the materials studied and the treatment applied before and during ageing are described, as well as the tests conducted for their analysis.

2.1. Materials

The insulating system was composed of paper and fluid. For the solid insulation, a commercial TUK paper with a thickness of 200 µm was used. This paper was combined with different dielectric fluids, including an uninhibited mineral oil, three natural esters sourced from various seeds (sunflower, rapeseed, and soybean), one modified natural ester from palm, and a synthetic ester. The key properties of these materials, as provided by their respective manufacturers, can be found in Tables 1 and 2. Comparing some values, the advantages of the alternative fluids, with respect to mineral oil, can be noticed, e.g., regarding their resistance to fire or their biodegradability as organic fluids derived from renewable sources. On the other hand, a possible drawback is their tendency to capture more water, but it seems not to be as harmful to the dielectric properties e.g. the breakdown voltage.

Table 1. Main properties of the TUK paper.

Property	Test Method	TUK
Density [g/cm^3]	IEC 60641	1
Tensile strength (machine direction) [MPa]	IEC 60641	115
Elongation (machine direction) [%]	IEC 60641	2
Moisture content [%]	ISO 287	<8
Electric strength in air [kV/mm]	IEC 60243	10
Electric strength in oil [kV/mm]	IEC 60243	70

Table 2. Properties of the studied commercial fluids.

Property	Mineral	Sunflower	Rapeseed	Soybean	Palm	Synthetic
Density 20 °C [g/cm^3]	0.839	0.91	0.92	0.92	0.97	0.839
Kinematic viscosity 40 °C [mm^2/s]	9.98	39.2	37	34	5.062	29
Flash point [°C]	176	330	>315	320	188	260
Fire point [°C]	-	362	>350	350	206	316
Pour point [°C]	−48	−25	-31	−18	−37.5	−56
Acidity [mg KOH/g]	<0.01	0.05	≤0.04	<0.05	<0.01	<0.03
Moisture [mg/kg]	15	150	50	4-50	52	50
Dielectric dissipation factor 90 °C	0.002	0.03	<0.03	<0.03	<0.003	<0.008
Breakdown voltage [kV]	46	65	>75	≥55	85	>75
Biodegradability	-	85	98	> 99	77	89

2.1.1. Mineral Oils

Mineral oil is derived from petroleum through refining processes, which are complex, resulting in slightly different compounds that may even differ when coming from the same manufacturer [26]. This oil is a mixture of aromatic compounds involving carbon and hydrogen atoms, essentially grouped into three classes of hydrocarbons in different proportions: paraffins (C_nH_{2n+2}), naphthenes (C_nH_{2n}), and aromatics (C_nH_{2n-6}), where n = 11–30 [27,28]. The proportion of each of these hydrocarbons will define the type of oil, which can be paraffinic or naphthenic [26]. The paraffinic structure can be a linear chain of carbon atoms, known as paraffin, or a linear chain with lateral branches, called isoparaffin. Naphthenes, on the other hand, are ring structures, typically formed by five, six, or seven carbon atoms, while aromatic compounds have double bonds between some of the carbon atoms. The petroleum refining process reduces the content of aromatic and polycyclic compounds, improving the dielectric strength and stability of the mineral oil [29]. The mineral oil used in this work is of the non-inhibited paraffinic type.

2.1.2. Natural Esters

These fluids are natural compounds synthesized from alcohols and fatty acids obtained from different plants. Their composition consists of triglycerides synthesized by esterification of glycerol with three fatty acids, which can be the same or different [26,30], and compounds with linear hydrocarbon chains terminated by the -COOH group. The chains can be saturated or mono-, di-, and tri-unsaturated, and the proportion of each type will vary depending on the type of plants used for the production of the esters. The presence of oxygen in the chemical structure supposes the existence of polar groups capable of linking water molecules, explaining the larger concentration of water in these fluids. This can be appreciated in their typical moisture content, which is shown in Table 2.

2.1.3. Synthetic Esters

Synthetic esters are chemical compounds obtained through an esterification reaction of an alcohol molecule with a carboxylic acid [26]. Depending on the alcohols and acids employed, different synthetics can be obtained. Again, the oxygen explains its hygroscopic capacity.

2.2. Experimental Procedure

2.2.1. Treatment

The fluids and paper were dried independently. Oil was dried in a vacuum oven at 60 °C for 24 h by alternating 4 h under vacuum (10 mbar) and 1 h in a nitrogen atmosphere (600 mbar). The paper was dried inside ageing vials, which were made of glass and had a capacity of 125 mL. This process was carried out in an air-circulating oven at 105 °C for 3 h. Moisture contents of the fluids and paper after drying, measured using the Karl Fischer volumetric titration following the IEC 60814 standard [31], are outlined in Table 3, all of which fall within the limits specified by IEEE [32].

Table 3. Moisture contents of the insulating materials after the drying process.

Moisture	Mineral	Sunflower	Rapeseed	Soybean	Palm	Synthetic
Fluid [ppm]	25.3	92.8	40.8	72.1	64.9	107.2
TUK [%]	0.5	0.9	0.6	1	0.9	1

Then, the vials were filled with 100 mL of oil, and copper plates were introduced into them, as shown in Figure 1. A consistent oil/paper/copper mass ratio of 16.3:1:1.3 was maintained in all vials, following the procedure outlined in the IEEE Standard C57.100 [33]. Finally, the vials were sealed with a PTFE/butyl septum and an aluminium cap in an air atmosphere. Before subjecting them to the ageing temperature, the vials were placed in a temperature-controlled oven at 60 °C for 2 h to ensure that the paper was fully impregnated by the fluid.

(a) (b)

Figure 1. (**a**) Ageing test materials and (**b**) Prepared vial.

After that, samples of each fluid were taken for the initial point analysis. The remaining samples were placed in the ageing oven, where the temperature was set to 150 °C, for a six-week ageing process and analysing the samples at six intervals: 16, 40, 70, 168, 384, and 1008 h.

2.2.2. Characterisation

The performance of both the oils and paper depends on various properties, which were measured at different ageing states to quantify degradation. Tests were conducted following applicable standards.

Some critical dielectric properties were determined using a BAUR DTA 100C dielectric oil analyser. AC BDV was measured at 1 kV accuracy, following the IEC 60156 standard [34]. Moreover, tan δ, relative permittivity (ε), and resistivity were measured using a BAUR DTL

C. Parameters were determined at 90 °C under AC sinusoidal voltage for tan δ and ε and under DC power for resistivity, following IEC 60247 [35].

Regarding the physical properties, parameters affecting cooling capacity, such as density and viscosity, were measured using a Stabinger Viscometer SVM 3000 at 15 and 40 °C. On the other hand, the flash and fire points were measured following ASTM D92 [36], using OilLab 670 equipment. Finally, the interfacial tension was measured with a force tensiometer K11 using the ring method at room temperature, as described in IEC 62,961 [37].

With respect to the chemical properties, the acidity was measured through potentiometric titration according to IEC 62021-1 [38] for the mineral oil and IEC 62021-3 [39] for the esters. The moisture content was controlled especially for the dielectric tests, following the IEC 60814 [31].

The DP of the paper was determined following ASTM D4243 [40]. The paper was crumbled and dissolved while being stirred for 16 h. Then, the viscosity of the resulting solution was measured and related to the DP of the paper sample.

3. Results and Discussion

In this section, the evolution of properties during material degradation is presented. The order in which they are shown has been defined based on the impact of ageing.

3.1. Parameters Directly Affected by the Ageing

This section includes parameters whose changes are directly affected by ageing and are not dependent on the evolution of other properties. For fluids, these include moisture content and acidity; for paper, these involve moisture content and polymerisation degree (DP). It is also important to clarify that variations in these parameters are the main drivers of changes in other properties, which will be addressed later in Section 3.2.

3.1.1. Moisture Content

Moisture, both in solid dielectrics and liquids, is a key factor during the ageing of insulating materials, as a high moisture content in paper usually implies rapid degradation [41]. The evolution of moisture along ageing time in the insulating systems is shown in Figure 2, which represents the moisture of the paper (%) by bars and that of the fluids (ppm) by lines at different stages in the ageing process.

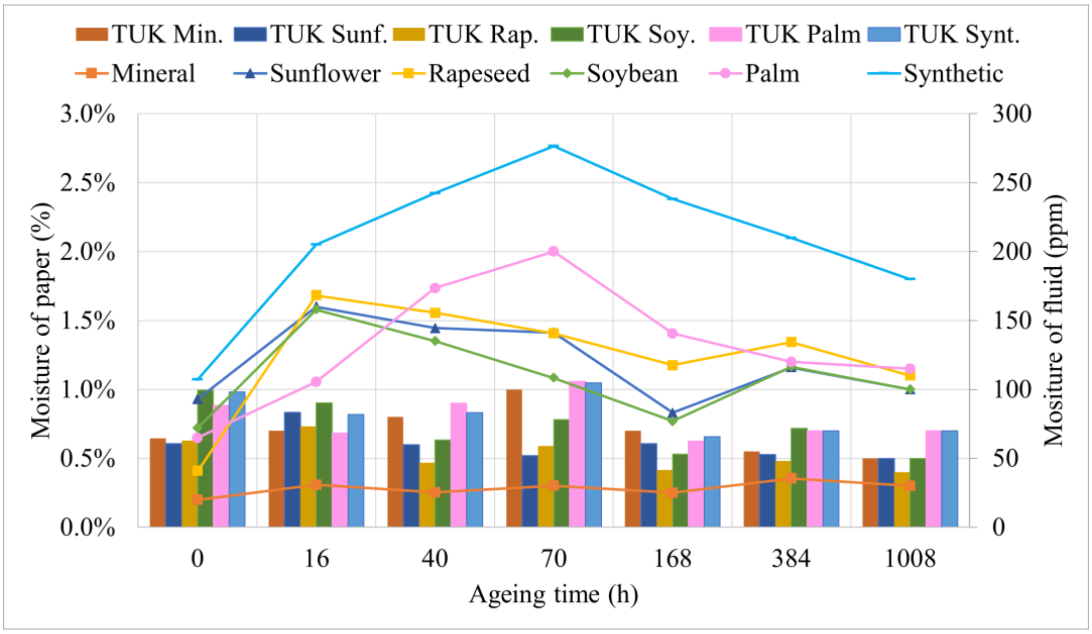

Figure 2. Evolution of the moisture content of the insulating system.

In the case of mineral oil, its moisture remained nearly constant, varying between 20 and 30 ppm, as also found in [42,43]. Its paper samples reflect a larger variation in their moisture, which is almost doubled during the first hours. In contrast, the esters exhibited a very different behaviour. All of them experienced an initial increase in moisture in the fluids, from less than 90 ppm to 160 ppm for the unmodified natural esters, from 65 ppm to 200 ppm in the palm ester, and from 107 ppm to 277 ppm in the synthetic one. After that, the moisture content decreased. Regarding the paper samples, the increases were lower than with the mineral oil, which also tends to be reduced with time.

Changes in moisture content during ageing are due to water being a byproduct of degradation caused by oxidation and paper hydrolysis [25,44]. In the beginning, there was an increase in moisture in the fluid–paper system, due to significant degradation during the first hours of ageing [21,45]. In the case of esters, their hydrophilic nature caused water to migrate from the paper to the fluid, while in mineral oil, water remained in the solid [46,47]. Simultaneously, part of the water was consumed in the paper through thermal hydrolysis [48]. Hydrolysis also occurred in the esters, since the water reacts with their triglycerides [27,49]. This led to the final decrease in moisture in those fluids and papers where the water reacts once the degradation and water production slows down. In the palm and synthetic esters, it is more complicated for this reaction to occur, as they are more chemically stable fluids [50], resulting in a higher moisture content.

The differences in the maximum values between the esters are related to the fact that the moisture in the unmodified esters started to decrease at the beginning of the ageing. It continues to increase for a longer time in the other esters due to their lower hydrolysis, as was observed in [17,28,42]. This can also point to a higher degradation rate of paper in these fluids. The lower level of water in the mineral oil does not necessarily mean the same.

Comparing these results with those obtained for the conventional Kraft paper in a previous study [51], the same trends were observed. Although, it was found that the moisture content was lower in the vials with TUK paper. When aged with Kraft paper, the unmodified natural esters reached a moisture content of 300 ppm, whereas the maximum value with TUK paper was 160 ppm. This probably happened due to a higher degradation of the Kraft paper, which led to a greater amount of water generated in comparison with the TUK one.

3.1.2. Acidity

The evolution of the acidity is also a parameter commonly used as an indicator of ageing, both of the fluids and paper [52]. Each fluid has a typical value of acidity range when new; esters, except for the palm one, generally have higher acidity than mineral oil, attributed to their chemical structure composed of triglycerides [42]. Hydrolysis and oxidation reactions, both in esters and in paper, led to acid generation [27,44,48,53], which is reflected in the acidity of the fluids. At the same time, acids can act as catalysts for paper hydrolysis, known as acid hydrolysis [54], increasing its degradation.

The acidity results obtained in this work are represented in Figure 3, which illustrates the evolution of the acidity (mg KOH/g) of each fluid during the ageing process. When fluids were new, they exhibited low acidity, in all cases lower than 0.1 mg KOH/g [50]. During ageing, the acidity of all fluids increases, although not in the same proportion. The mineral oil showed the smallest increase in acidity (500%). With the palm ester, a relatively low increase in acidity (1100%) was detected, similar to the synthetic ester (900%), compared to the unmodified natural esters, where the increase was higher, up to 1700%, as shown in previous studies [18,50]. According to this, a direct relationship between acidity and degradation in the insulating system could be expected.

The acids detected in mineral oil mainly came from general oxidation and paper hydrolysis, as this oil did not react with water, as explained in [29,53]. With the palm and the synthetic esters, similar to mineral oil, acidity mainly came from oxidation and cellulose hydrolysis, as these fluids exhibit high chemical stability. The fact that these fluids suffer less hydrolysis conforms to their lower decrease in water, as seen in the previous section and justifies their lower acidity. In the last ones, the unmodified esters, in addition to the paper degradation, fluid oxidation, and hydrolysis, were relevant, causing larger acidities.

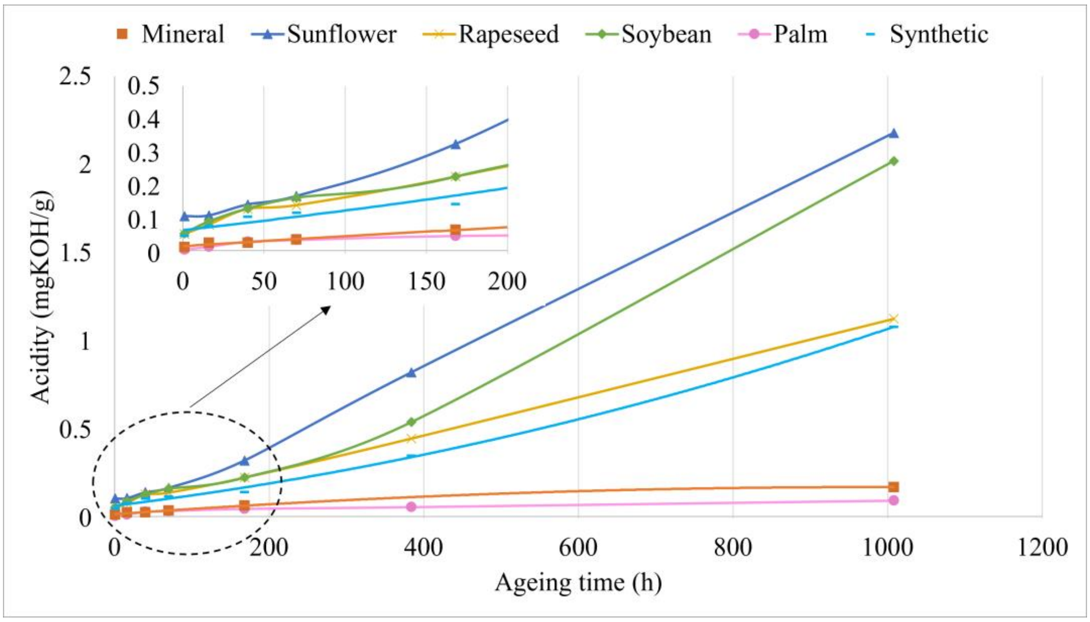

Figure 3. Evolution of the acidity of the dielectric fluids.

As it was mentioned before, the acids can even increase the degradation of the paper. However, it is important to explain that this depends on the type of acid generated, including high and low molecular weight acids, and on the chain length. Longer acid chains have higher molecular weights [49]. High molecular weight acids are formed in all dielectric fluids and papers, while low molecular weight acids appear in mineral oils [53]. It has been demonstrated that low molecular weight acids, due to their polarity and hydrophilic nature, tend to dissolve in the paper [55]. Consequently, they can react with cellulose, catalysing hydrolysis, which leads to the breakdown of cellulose chains and, consequently, material degradation. On the other hand, high molecular weight acids, generated in large quantities in natural esters undergoing hydrolysis, do not produce ions that can react with cellulose through hydrolysis. Contrarily, some authors [21,42,49] and even standards [53] suggest the possibility of a transesterification reaction between these heavy acids and cellulose. Through this process, acids would align parallel to the cellulose chain, binding to its OH groups, and forming a barrier against water and paper hydrolysis [56,57]. Therefore, the esterification of this group would reduce the hydrolysis suffered by cellulose and, consequently, its degradation and acid generation. Based on this, despite the higher acidity of the esters, they would not damage the paper any more than mineral oil and could even protect it.

Some differences were observed among the different unmodified natural esters in terms of acid formation. Rapeseed fluid exhibited lower acidity than the other esters, which showed similar levels among themselves. Supposing that hydrolysis is similar among them, based on their common chemical structure, which implies similar acid generation, the difference in the acidity could be due to less paper degradation when aged with this ester. Additionally, a more significant transesterification may also explain this behaviour if it is considered, as it supposes not only a reduction in the degradation but also a reduction in the acids in the fluid, as they keep linked in the cellulose.

Comparing the acidity of the same fluids when aged with TUK and Kraft paper, shown in [51], it was found that the acidity is lower when aged with the TUK one. This could be explained by the higher thermal resistance of this paper, whose treatment particularly protects it against hydrolysis [24,25], thereby reducing this reaction and consequently generating fewer acids.

3.1.3. Polymerisation Degree

The degradation of the cellulose could be analysed through different parameters, with DP being one of the most common. This property measures the amount of monomers in the cellulose polymer, which are reduced as a result of the hydrolysis, oxidation, and, in case of very high temperatures, pyrolysis [25,44,48]. The evolution in the DP in the TUK paper analysed in this work is shown in Figure 4. This figure represents how the integrity of the paper decreases the longer it is subjected to the test temperature.

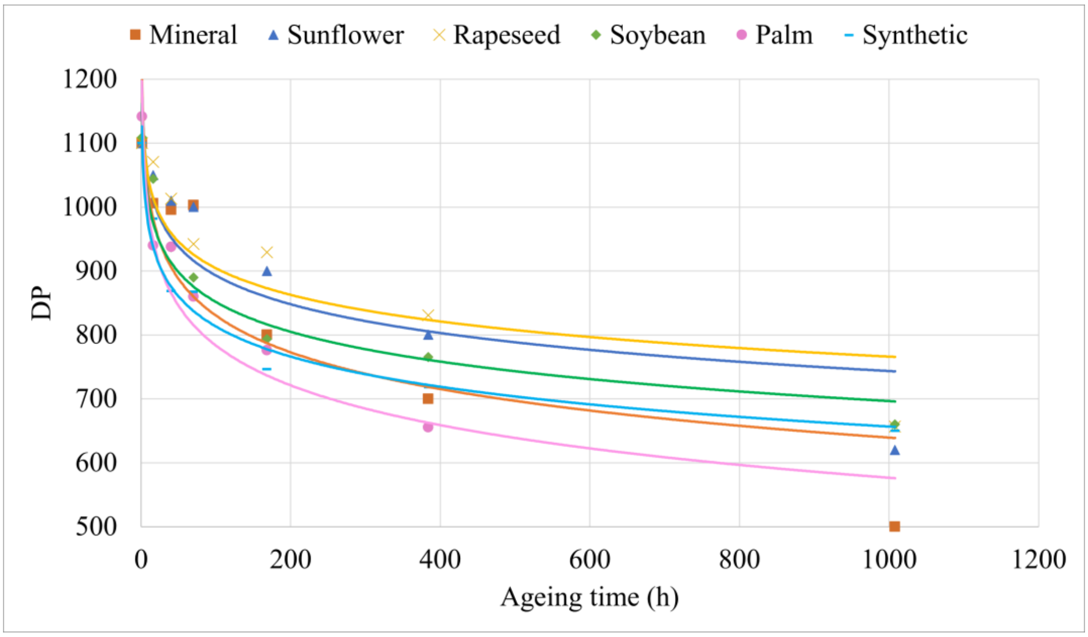

Figure 4. Evolution of the DP of the TUK paper.

As observed in the obtained results, DP decreased rapidly at the beginning of ageing, slowing down the degradation rate with a logarithmic trend, as shown in other studies [44]. This meets with the evolution in the moisture content of the fluids, with a fast increase in the first stages of the tests.

The DP decreased in all the unmodified natural esters at a similar rate and to similar levels. These fluids demonstrate better protection for the paper compared to other fluids as the DP values are higher with them. The slightly better performance of the rapeseed fluid with respect to the sunflower and soybean esters could be related to the lower acidity, as a result of a lower degradation of the paper. In tests with synthetic ester, the achieved DP was lower than with the previous fluids but better than with palm and mineral oil. In fact, the TUK paper aged with palm ester showed the highest degradation. The superior performance of unmodified natural esters in the TUK paper degradation could be justified by the hydrolysis occurring in these fluids and the presence of high molecular weight acids and the transesterification [21,42]. However, the greater degradation of this paper in palm ester cannot be attributed to significant changes in its properties and finding a specific cause for this behaviour proves challenging but could be caused by severe hydrolysis of the paper, since a white substance appeared in those vials.

As the ageing progresses, the reduction in the DP is less pronounced, as the degradation is reduced. This means a lower production of water, which supports the change in the evolution of the moisture in fluids and papers if hydrolysis is also considered. Here, it is also noticeable how the lower level of the moisture content or acidity in the mineral oil is not directly related to the integrity of the papers impregnated. In fact, in this fluid, the paper's degradation is larger than in most of the esters. It is also related to the higher content of water in its paper, which is known as a promoter of paper degradation by hydrolysis. As mentioned in the discussion of the moisture content, the evolution seen with the palm and the synthetic esters may point to a higher paper degradation, as confirmed by these DP results.

In comparison with the Kraft paper [51], the TUK paper degradation was noticeably lower for the same ageing period, as was found in other works [45]. This meets with the trends seen when comparing the moisture content and the acidity. The main difference between both papers was related to the performance of the palm ester, which was better than the mineral oil with the Kraft paper and the worst fluid with the TUK one.

3.2. Parameters Affected by the Ageing and by the Changes in Other Properties

Whilst the aforementioned parameters were directly affected by the hydrolysis and oxidation, there is another group of properties whose variations are also influenced by the changes occurring in the former. These properties, including the dielectric properties (breakdown voltage, dissipation factor, and resistivity) and the interfacial tension, are mainly modified by changes in moisture and acidity of the fluids, as an indirect consequence of ageing.

3.2.1. Breakdown Voltage

The BDV is affected by the presence of dissolved or free water in the fluid, as well as solid contaminants. Both types of compounds tend to migrate to areas with high dielectric requirements, i.e., areas of electrical stress, leading to a reduction in the BDV [58]. Due to the great volume of sample needed for this measurement, the BDV was only measured at three ageing states, as shown in Figure 5.

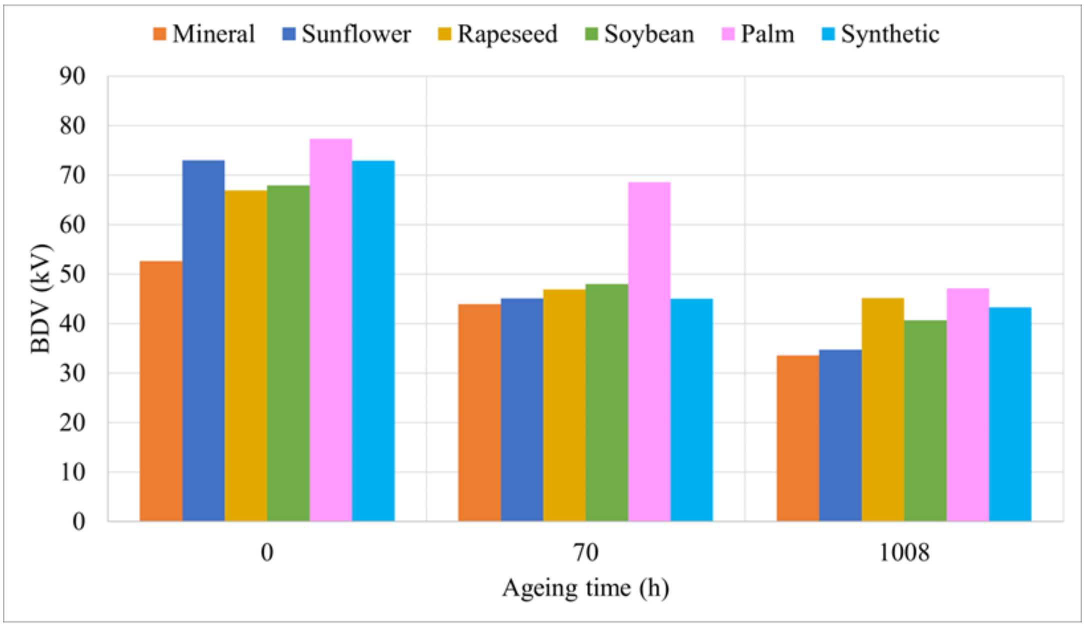

Figure 5. Evolution of the BDV of the dielectric fluids.

During the ageing, the BDV decreased in all fluids. The decrease was more abrupt in natural and synthetic esters than in mineral oil, leading to a significant drop in dielectric rigidity during the initial ageing period (up to 40% in sunflower ester). During the second half of the test, which has a longer duration, a very small reduction occurs, up to 20%, and in some cases, non-existent. This variation pattern could be caused by the increase in the moisture content at the beginning of the ageing, which impacts the dielectric capacity. Towards the end of the ageing, the moisture decreased or remained constant. Thus, the moisture at the last test was lower than at the intermediate one, which would explain the slight reduction in the BDV, since the greater degradation of the fluid could be offset by the decrease in water content. In fact, some authors concluded that BDV, although being a property with significant variability in its measurement [59], is more affected by moisture than by fluid ageing [17,45]. On the contrary, in the mineral oil, the decrease in the BDV was more gradual and detected in both periods, being higher in the second part of the ageing (−16% after 70 h, and −22% between 70 and 1008 h of ageing). The moisture of this fluid was practically constant in all tests, with no significant differences observed between any of the analysed points. Therefore, the reduction in the BDV could only be attributed to oil degradation and the presence of other types of byproducts or contaminants but not to changes in the water content. This could explain why variations as abrupt as those experienced by alternative fluids were not detected.

3.2.2. Dielectric Dissipation Factor

The tan δ is one of the most commonly used properties in the analysis of the dielectric condition of liquid insulation. It depends on both the polarity and conductivity of the fluid, with higher values observed when these parameters are greater [35]. The evolution of tan δ of each fluid throughout the ageing is represented in Figure 6, which shows the increase in the dielectric losses as the fluid degrades.

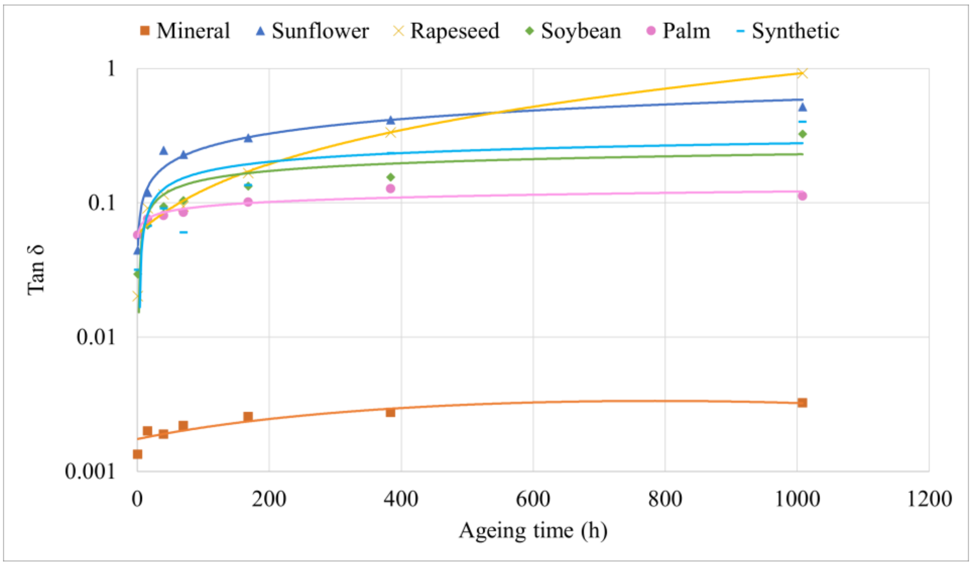

Figure 6. Evolution of the tan δ of the dielectric fluids.

As seen in the properties provided by the manufacturer, Table 2, and in accordance with the results obtained by other authors [43,60], the esters had a higher tan δ than mineral oil. This is because their polarity and conductivity are higher than those of traditional oil [50].

During the ageing, the tan δ increased in all the fluids, following an exponential trend, with the fastest increase occurring at the beginning of ageing, followed by a nearly linear increment, except for the rapeseed fluid. The increase in tan δ was very low in the mineral oil and palm ester, with an increment of approximately 100%, whereas in the sunflower, soybean, and synthetic esters the increase was approximately 1000%. In the case of the rapeseed ester, it was clearly affected by the ageing, with an increment of 4500%.

The variation in tan δ can be caused by different factors. The changes in the moisture content affect the resistivity of the fluid since water is a conductive particle and thus also affects its tan δ [61]. Additionally, any polar nature contaminant appearing in the fluid will have effects on this property. Moreover, acids present in the fluid can affect the loss factor [62], especially those of long chains [58]. Finally, some studies have also indicated that oxidation is a factor with significant influence on the tan δ of dielectric fluids due to the changes it causes in their structure [63]. Thus, even a small amount of oxidation can cause significant variations in dielectric losses. In the case of the rapeseed ester, its higher increase in the tan δ cannot be attributed to a higher moisture content or acidity, in comparison with the other esters. However, it could be due to a different length of the acids' chain, since the structure of the esters differs from one to another depending on the origin [12,13].

In comparison with the results obtained for Kraft paper in [51], it was found that the tan δ of all the fluids was higher when aged with TUK paper, despite the lower acidity of the fluids and the significantly lower ageing of the paper. This could be attributed to the polar compounds of the TUK paper due to the treatment applied to improve its thermal performance, which increases the nitrogen content [24,25]. The use of this paper led to an increase in the dielectric losses of the fluids, but it did not affect all the fluids equally. Palm ester was the fluid least affected by the use of TUK paper, along with mineral oil. The natural and synthetic esters were notably and similarly affected, with the effect being particularly pronounced in the case of the rapeseed one. One of the reasons for this different behaviour could be due to differences in the compositions of the fluids, which could lead to varying degrees of attraction to these polar substances.

3.2.3. Resistivity

Resistivity results versus time are collected in Figure 7.

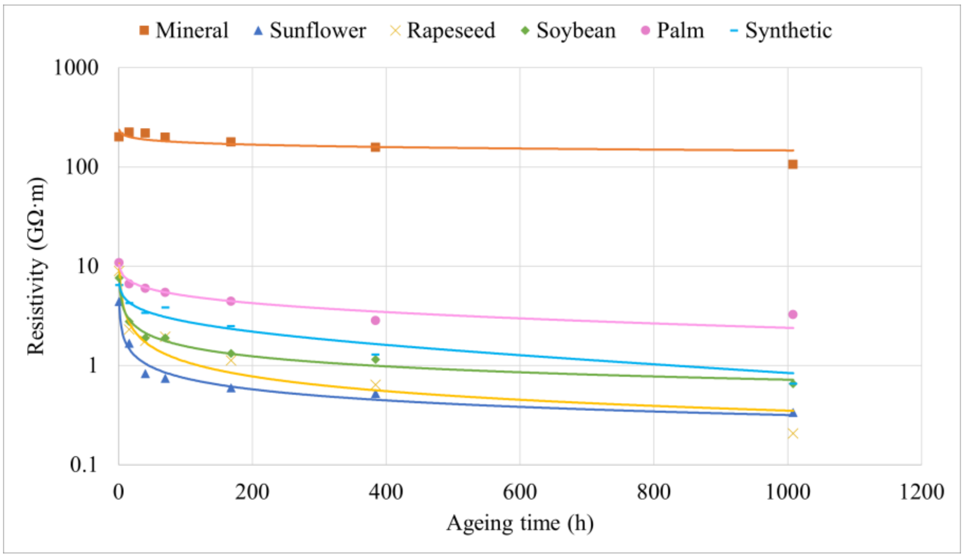

Figure 7. Evolution of the resistivity of the dielectric fluids.

The mineral oil had the highest resistivity during all the experiments, being 60 times higher than that of the alternative fluids, as it was also found in other works [19,64]. Regarding the esters, slight differences were found between them.

The resistivity decreased with the ageing, as a result of the changes in water content and other contaminants, as explained in the previous section. Despite tan δ also depending on this parameter, a different behaviour of the rapeseed fluid with respect to the other esters in terms of resistivity was not found, so it seems that the rapeseed fluid was affected by an increase in its polarity.

Comparing the results with those obtained for the Kraft paper in [51], it was found that the resistivity was slightly higher with that paper than with the TUK one, but the effect of the paper was less significant than in the tan δ.

3.2.4. Interfacial Tension

This parameter depends mainly on the polarity of the fluid or the presence of polar substances or contaminants in it [63]. Due to the chemical structure of the esters, their polarity is higher than that of the mineral oil [65]. The interfacial tension results at different times are represented in Figure 8—the higher the polarity, the lower the interfacial tension.

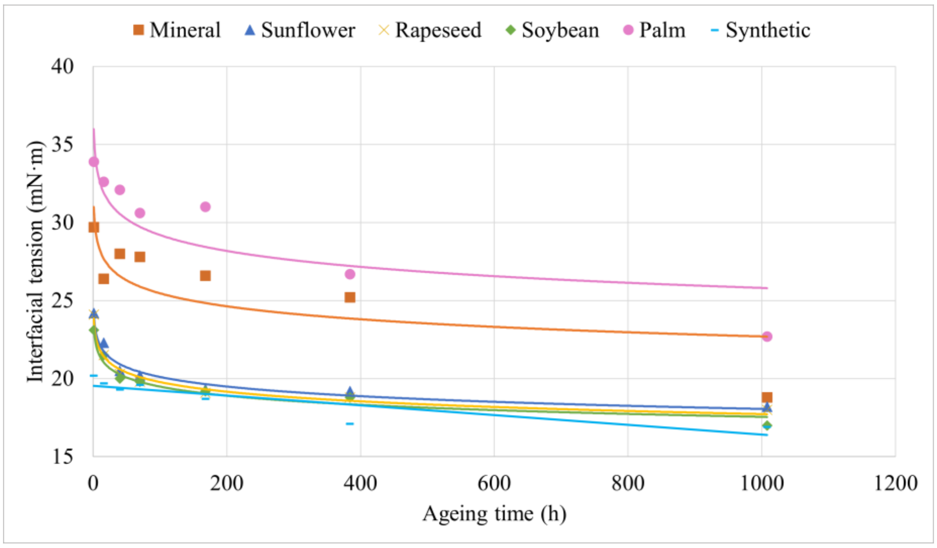

Figure 8. Evolution of the interfacial tension of the dielectric fluids.

The interfacial tension decreased with the ageing, especially at the beginning, since polar substances are generated, and then it tended to stabilize as a result of the decrease in the degradation rate, as was also observed in [14,66]. Despite the lower interfacial tension of the mineral oil with respect to most of the esters, this parameter decreased the most in this fluid, by approximately 40% at the end of the experiment, while in the esters, this reduction was lower than 20%. The difference in the changes in the interfacial tension between the mineral oil and the esters could be related to the acidity. Despite the lower acidity of the mineral oil, it was the only fluid in which low molecular weight acids, that are polar, could be generated, [25].

Moreover, in comparison with the Kraft paper results [51], it was found that the interfacial tension is lower when the fluids are combined with the TUK paper. This agrees with the increase in tan δ previously explained, since the decrease in the interfacial tension is also related to the increase in the polarity.

3.3. Parameters Not Affected by the Ageing

Some of the analysed properties did not undergo changes due to ageing. The unaffected properties are primarily of a physical nature, including density, viscosity, and flash and ignition points, as well as permittivity, which is a dielectric factor.

3.3.1. Density and Viscosity

Any variation in the density of the fluid is crucial to the cooling of the transformer since it affects the natural convection. However, this property remained constant with the ageing, according to the experimental results measured for each ageing state, which are summarized in Table 4 for all the fluids.

Table 4. Density of the dielectric fluids [g/cm^3] at 15 °C.

Mineral	Sunflower	Rapeseed	Soybean	Palm	Synthetic
0.861	0.917	0.922	0.924	0.863	0.971

On the other hand, viscosity is the property that most affects the cooling capacity of the fluid, since it determines the speed of the fluid and its movement inside the transformer tank. The unmodified natural esters are characterised by having high viscosity, much higher than that of mineral oil, which is one of the major drawbacks of these fluids. On the other hand, the main advantage of the palm ester is its low viscosity. The viscosity results measured at three ageing states are represented in Figure 9. This property exhibited just a slight variation with degradation in this study, since it mainly depends on oxidation and it is not relevant in sealed systems, as demonstrated in [27,58]. Because of that, the viscosity is nearly constant, with a variation of less than 1%, which was also found in other studies [20,67].

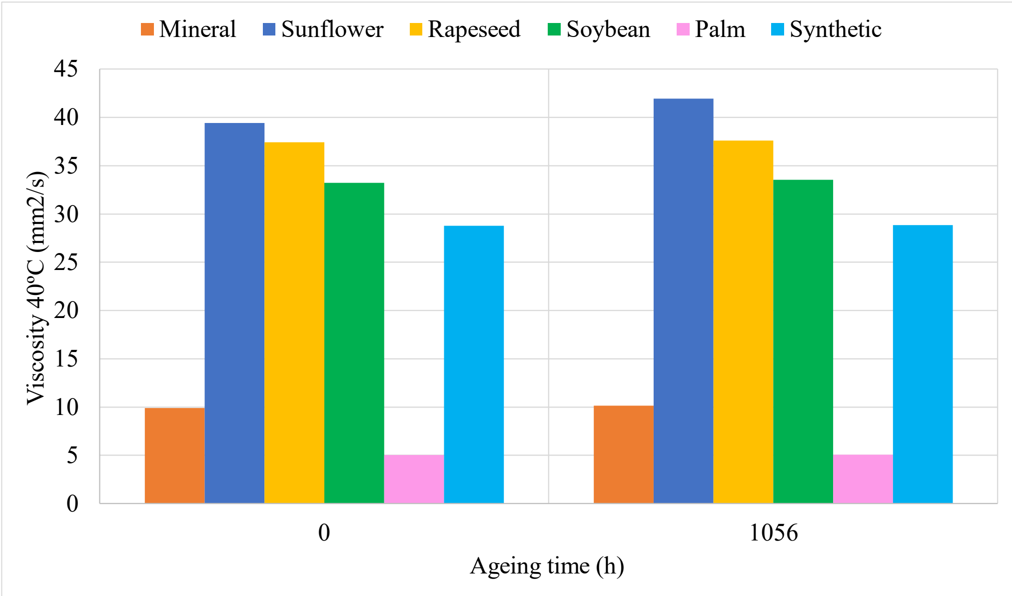

Figure 9. Evolution of the viscosity of the dielectric fluids.

3.3.2. Flash and Fire Points

The risk of fire depends on the maximum temperature that the dielectric materials can withstand. In Figure 10, the results of the flash and fire points of each fluid are depicted as solid and dashed lines, respectively, both for the initial state and for the final state of ageing.

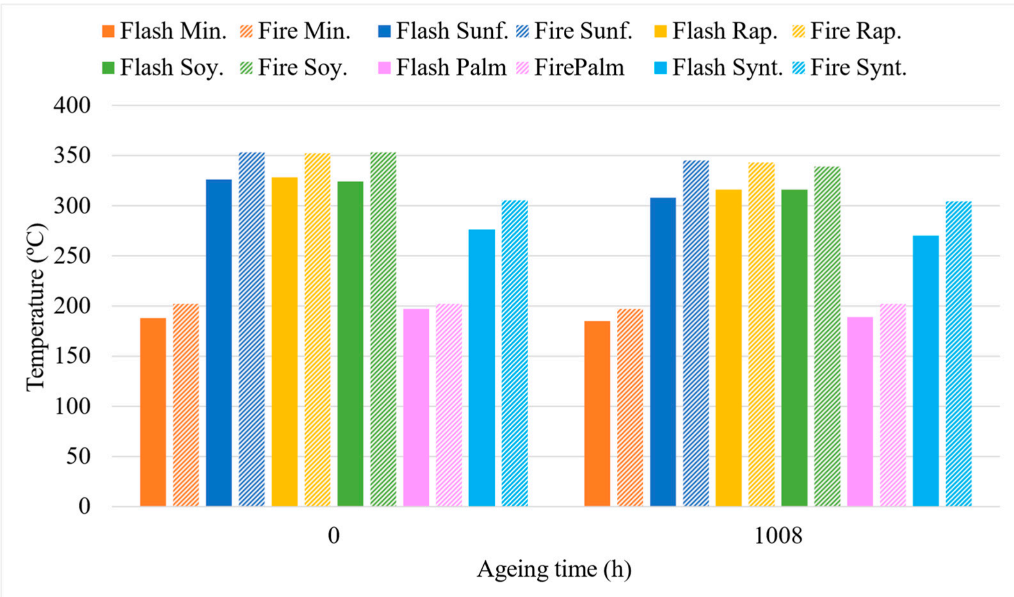

Figure 10. Evolution of the flash and fire points of the dielectric fluids.

As can be seen, all the unmodified natural esters had the highest flash and fire points, whereas the palm ester withstood lower temperatures, similar to those of the mineral oil. The fire points were approximately 30 °C higher than the flash points in the natural and synthetic esters, whereas they were only 10 °C higher in the mineral oil and palm ester. These parameters did not change with the degradation of the fluid. Moreover, if comparing the results with those obtained previously [51], they show that the cellulosic material did not affect these properties.

3.3.3. Dielectric Permittivity

Finally, the dielectric permittivity is important in the dielectric design of transformers, as the relationship between the permittivity of the fluid and paper will largely determine the distribution of the electric field throughout the insulating system [68]. This determines the electrical stress the materials withstand and the occurrence of partial discharges. Figure 11 shows the relative permittivity of each fluid at the initial state and at the end of the ageing. According to these results, the relative permittivity of esters is significantly higher than that of mineral oil, approximately 1.3 times that of the conventional fluid and more similar to that of paper, which has a permittivity of approximately 3 [69,70]. This parameter underwent minimal variations during ageing, as observed in other studies [19,43]. Also, considering the results of [51], the permittivity is not affected by the insulating paper with which the fluid is combined.

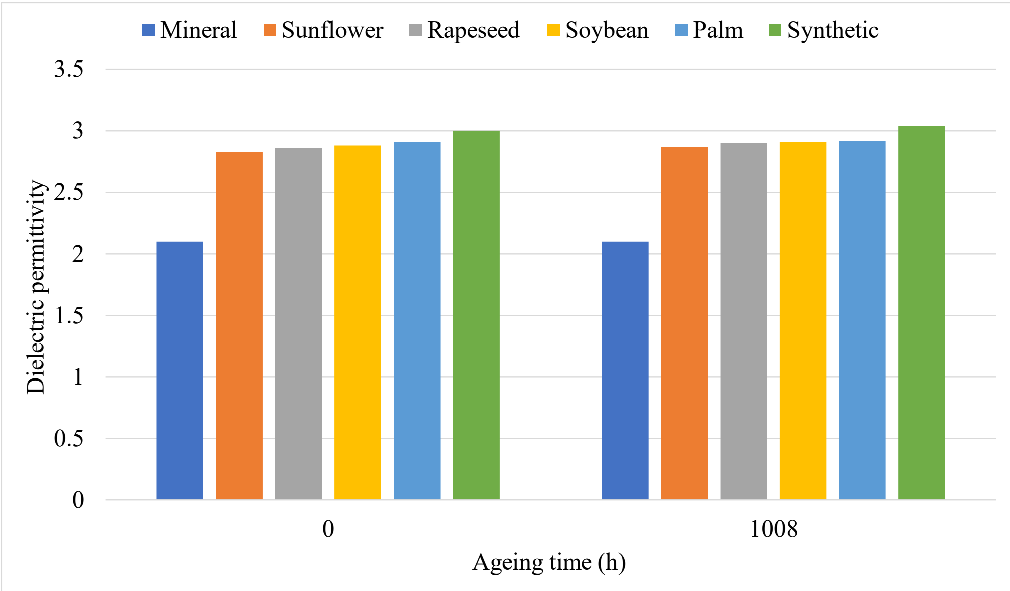

Figure 11. Evolution of the dielectric permittivity of the dielectric fluids.

4. Conclusions

The assessment of the main properties of the top five sustainable alternative fluids to mineral oil and their evolution have been studied under the same conditions, making it possible to compare them. The characterisation included the main parameters that determine the suitability of a fluid for use in transformers.

It has been found that both unmodified natural esters and synthetic esters have very different properties from mineral oil, initially appearing worse, such as high acidity, dielectric loss factor, and viscosity. Among them, there are very slight differences, but they all are within the same order of magnitude and comply with their respective regulations. On the contrary, the palm ester is a unique case, its properties are very similar to mineral oil, making it theoretically more suitable for use in transformers. However, it is important not only to evaluate the initial properties of the fluids but also their overall evolution and their interaction with the paper.

It was found that some properties were affected directly by the ageing, such as the moisture content, acidity, and DP of the paper, whereas other properties changed as a result of the changes in the previous ones, such as BDV, tan δ, resistivity, and interfacial tension. It was also detected that the other properties, mainly physical, were not affected by the ageing, including the density, viscosity, and flash and fire points and that the permittivity did not change.

From the results, it can be drawn that the similar behaviour of the three unmodified naturals makes them equally good alternatives for use in transformers. Although their properties are slightly different, their evolution is alike, with none standing out for greater degradation of the paper. Indeed, the TUK paper is better protected by the unmodified natural esters, followed by the synthetic fluid, with one exception. Only the palm fluid aged faster than the mineral oil, despite the fact that, based on its dielectric and physicochemical properties and the evolution of them, it seemed to be the fluid with the best performance. On the whole, if these alternative fluids were used, the disadvantages that may arise, such as increased heating, are offset by improved temperature resistance, while the enhancement in sustainability remains.

From the comparison with previous work, it was observed not only that the TUK has greater resistance to ageing than conventional Kraft paper but also that the better performance of most of these esters does not depend on the type of paper. A practical application of these fluids will result in better protection of the cellulosic solid insulation that conditions the lifespan of transformers, which means a prolongation of their years of exploitation. However, the use of the upgraded solid increases the polarity of the fluids, negatively affecting dielectric losses, especially in the rapeseed ester.

As a main conclusion, it could be said that no significant differences have been found among the alternative fluids to determine that any of them is better, except for the degradation of the TUK paper with the palm ester.

Based on this, the use of alternative biodegradable fluids can enhance the sustainability of the electrical system in two ways. On the one hand, they enable the reduction in fossil raw material usage by being derived from renewable resources. On the other hand, by protecting the insulation paper from ageing, they extend the equipment's lifespan, both in new and operational power transformers, thereby reducing the consumption of resources for manufacturing new machines.

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