New Insulating and cooling liquids for transformers

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Abstract:
The combination of a solid and a liquid insulation is the most frequently used insulating system in high voltage apparatus, where components have to be insulated and loss heat has to be dissipated. The requirements on the liquid part of the insulating system are not only the electric and dielectric performance but also the performance regarding environmental requirements and dehydration capability as well as low inflammability. The use of ester liquid, partly or totally replacing mineral oil, reduces the risk of environmental pollution, increases the lifetime of the component and reduces the fire risk. Furthermore the capability of ester liquid to assimilate a higher amount of water in comparison to mineral oil without reducing its electric strength reduces the breakdown endangerment during cold start and allows the operation of the component at higher temperatures.

The contribution presents a comparison of the performance of ester liquid and mineral oil as well as oil/ester mixtures concerning the electric behaviour and covers the whole range of blends between ester liquid and mineral oil. The dielectric parameters like dissipation factor and permittivity have been measured for the whole mixture range in order to evaluate the performance of the complete insulating system regarding the compatibility of the liquid insulation with different types of paper.

1. Introduction
Because of the wide availability and low cost, mineral-based transformer oils are probably the most widely used electrical insulating liquids in the world today - and have been for the past century [1]. Between 1930 and the mid seventies, non-flammable liquids like PCB (some trade names included Askarel, Arcolors, Pyranol, Inerteen, Chlorextol, etc.) were used for insulation purposes. During the 1970s it was determined, that PCBs can be an environmental hazard and can accumulate in the environment. Especially in case of a surrounding fire PCBs can generate high levels of dioxins and furanes. Since that time researchers tried to find substitutes for PCBs in transformers.

Since then transformers have been filled with different synthetic liquids such as silicone, ester, perchlorethylene etc. They have good dielectric, heat transfer and aging properties, but because of their relatively high
cost their use has been limited to special transformer applications [2]. One of the major advantage, forcing the application of ester liquids, is their improved environmental compatibility. Additionally these liquids comprise several extra advantages, which are at first their lower inflammability. Transformers can contain several tons of insulating liquid, that may cause a long lasting pool fire in case of a transformer tank rupture due to winding failures. Ester liquid and mineral oil are of an almost similar density. They are completely miscible at any ratio. Almost all electrical and dielectrical properties of ester liquids are similar to mineral oils despite the relative permittivity $\varepsilon_r$, which is higher than those of mineral oils. This is an additional benefit, if the ester oil is used for impregnating cellulose, as the relative permittivity of the liquid is closer to the one of cellulose (about 5.6), thus resulting in a more uniform electrical field distribution within the combined insulation. Some of the physical and electrical properties of mineral based transformer oil and ester liquids are summarised in Table 1. Actually the use of ester liquids in larger units is under discussion and some transformers up to a power of several tens of MVA are in operation filled with them.

<table>
<thead>
<tr>
<th>Table 1: Properties of the investigated insulating liquids</th>
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<tbody>
<tr>
<td><strong>Unit</strong></td>
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<tr>
<td><strong>General physical properties</strong></td>
</tr>
<tr>
<td>Density at 23°C (kg/m³)</td>
</tr>
<tr>
<td>at 90°C</td>
</tr>
<tr>
<td>Pour point °C</td>
</tr>
<tr>
<td>Toxicity</td>
</tr>
<tr>
<td>Ability to biodegradability</td>
</tr>
<tr>
<td>Water solubility at 20°C ppm</td>
</tr>
<tr>
<td>at 100°C</td>
</tr>
<tr>
<td><strong>Heat transfer capability</strong></td>
</tr>
<tr>
<td>Cinematic viscosity at 20°C mm²/s</td>
</tr>
<tr>
<td>at 90°C</td>
</tr>
<tr>
<td>Calorific capacity at 20°C W/(m K)</td>
</tr>
<tr>
<td>at 90°C</td>
</tr>
<tr>
<td>Specific heat at 20 °C kJ/kg.K</td>
</tr>
<tr>
<td><strong>Fire properties</strong></td>
</tr>
<tr>
<td>Flash point °C</td>
</tr>
<tr>
<td>Flame point °C</td>
</tr>
<tr>
<td>Combustion heat kJ/kg 10⁻³</td>
</tr>
<tr>
<td>Self ignition temperature °C</td>
</tr>
<tr>
<td><strong>Electrical properties</strong> (all the data are relative to a temperature of 23°C)</td>
</tr>
<tr>
<td>Breakdown strength (ac) $U_{VDE}$ kV</td>
</tr>
<tr>
<td>Permittivity $\varepsilon_r$ (25°C, 50 Hz)</td>
</tr>
<tr>
<td>Dissipation factor tan δ (90°C)</td>
</tr>
<tr>
<td>Volume resistivity $\Omega$ cm</td>
</tr>
<tr>
<td>(*) The data are mean values obtained from many manufacturers.</td>
</tr>
</tbody>
</table>
The paper presents results of investigations on ester liquid and mixtures of ester with mineral oil. The investigated mixtures are a combination of a widely available mineral oil and a specific amount of ester liquid, which has similar electrical properties combined with fewer environmental risks, less fire hazards and high hygroscopicity. The water saturation limit of esters is more than 50 times higher than that of mineral oils. The investigations have been carried out on unaged mixed liquids as well as on specimens under severe aging conditions. Pure liquids have also been investigated to provide baseline data for comparison.

2. Description of the liquids

The investigations have been carried out using three different insulating liquids commonly used in high voltage transformers and a mixture of two of them recently proposed as insulating liquid for transformers [2]. A short description of them is presented hereafter.

2.1 Mineral oil

Mineral oil so called "transformer oil" is made by refining a fraction of the hydrocarbons collected during the distillation of a petroleum crude stock. The crude oil stocks and the refining processes used in producing these oils are typical of those used in producing many common petroleum lubricating oils. Chemically, the mineral oil consists of a complex mixture of basic hydrocarbon liquids such as paraffinic (40 to 60%), naphthenic (30 to 50%), aromatic (5 to 20%) and olefinic (roughly 1%) components. The chemical structure of them is described in the literature [4 - 7]. The early transformer oils were paraffin-based but, after about 1925, they have been replaced by napthenic oils because of the high pour point of paraffinic oils. It was soon recognized, that paraffinic oils are prone to sludging, and this resulted in the development of oil reclamation techniques [8]. Their relatively good aging behavior and low viscosity make mineral oil a good insulating and cooling liquid. Its electrical and dielectrical properties are strongly temperature and humidity dependent. The main advantages of this petroleum-based oil are its wide availability and its low cost. However mineral oils posses a low relative permittivity, a low flash point and are slightly toxic. Mineral oil posses also the disadvantage of endangering the environment in case of a transformer leakage [3].

When searching substitutes for PCB, ecological considerations made raise the problem of searching incombustible and non-toxic insulating liquids, thus ester liquids and silicone fluids have been proposed. They belong to the HFP (high fire point) liquids also known as „less inflammable“ liquids. It is to notice, that by definition a HFP liquids must have a minimum fire point of 300°C [4]. Their relatively high cost and availability has limited their use to special transformer application.

2.2 Ester Liquid

The ester liquid used for these investigations consists of Pentaerythritol-Tetraester and different fatty acids. The chemical structure of it can be found in the literature [4-7]. Ester liquids are non toxic, well digested by micro-organisms and posses low vapor pressure at operating temperatures of high voltage transformers. In fire they generate no dioxins or toxic products and possess a good biodegradability [9]. Previous work [3] has shown, that ester liquids can be used for the retrofilling of mineral oil filled transformers. These features, coupled with the ability of drying the solid insulation (impregnated paper), are considered as positive aspects. However, they are also proned to possible hydrolytic detachment through their high moisture saturation limit.

3. Relative Humidity
By definition, the relative humidity of a liquid is the dissolved water content of the liquid relative to the maximum capacity of moisture, that it can hold at specific temperature. The relative humidity $W_{rel} (%)$ at a given absolute temperature $T(K)$, is defined in terms of the absolute water content in the liquid $W_{abs}$ (ppm) versus the saturation limit $W_L(T)$ such as :

$$W_{rel} = \frac{W_{abs}}{W_L(T)}$$ (1)

The saturation limit of an insulating liquid depends on the type of insulating liquid, its chemical composition and its mean molar weight [7]. The temperature dependence of maximum water solubility of the insulating liquids is described by the following well-known exponential equation [7, 10]:

$$W_L = W_0 \exp\left(\frac{H}{T}\right)$$ (2)

where $W_0$ (ppm) and $H$ (K) are material constants, which have to be determined experimentally for each insulating liquid [4, 7, 11]. In a logarithmic diagram, equation (2) depicts a linear curve. Therefore, only two points are necessary to describe the insulating liquid behavior. For this reason, we only performed investigations at three different temperatures (20, 60, 88°C) and extrapolated the results from 0°C to 100°C. The material constant $W_0$ and $H$ of the investigated insulating liquids can be determined from table 1. Figure 1 shows the water saturation limit of the investigated liquids for an absolute temperature range of 0°C to 100°C. As illustrated, the water solubility of ester is much higher than those of mineral oil, followed by the mixture of mineral oil with 20% ester liquid and silicone fluid. The difference in the behavior of silicone and ester liquids compared to mineral oil can be explained by the difference in water absorption of these liquids. Ester or silicone liquid can take up water in chemically bounded and in dissolved form, while in mineral oil, water is only dissolved [7, 11]. Indeed, it is well known, that water contamination in liquid may be present in three different states, namely dissolved, emulsified and dispersed [6, 7, 11]. Moisture can accumulate chemically in the oxygen-containing liquid. Especially the ester molecule contains chemically bond oxygen at an extent of about 20% by weight [12] and water linkages of the polar side valences lead to a high water saturation limit of it [6, 13]. However, investigations in the field of chemical species diffusion would be very helpful in understanding the process.

![Fig. 1: Water saturation limit of different liquid versus temperature](image-url)
4. Miscibility of the liquids

Ester liquid and mineral oil are of almost similar density [5, 6]. The mineral oil and a specific amount of the ester liquid have been poured into a vessel and slowly mixed using a stirrer. To assess the degree of miscibility, the relative permittivity and the dissipation factor have been determined at three different positions of the vessel for three types of mixtures. The results show, that there is no significant difference regardless of the amount of ester added. In order to assess the temperature effect on the mixtures, the specimens have been heated to 90 °C, and even after few days there was no visible separation of the mixed liquids. We can therefore conclude, that the two liquids are miscible up to 50% proportion of the ester liquid.

5. Investigated Parameters

The aim of the investigations was to determine those physical, electrical, dielectric and aging properties, which, according to the IEC standards, are used to classify transformer oils as to guarantee their quality and life. Although all the properties listed [1, 2] are important, some have a special merit for the characterisation of an insulating liquid, especially those, that are likely to vary significantly with the oil purity and composition as well as with temperature and electric field. The most important properties are the electrical strength and viscosity, followed by dissipation factor (tan δ), water content and neutralisation number [3,7,12]. The permittivity, dissipation factor, and AC electric strength of both the pure and the mixed liquids and their viscosity were examined. The results are compared with the limits given in IEC and VDE standards. To simulate real working conditions as well as critical situations in transformers, the temperature has been varied between 0 and 100 °C along with variations in the water content of the fluid.

The water content varied between 10 and 40 ppm for the mineral oil, between 10 and 185 ppm for the mineral oil plus 10% ester liquid, between 10 and 365 ppm for the mineral oil plus 20% ester liquid and between 10 and 1120 ppm for mineral oil plus 50% ester liquid. Only the average values at all the humidities at each temperature are shown in the figures. To determine the water content of the insulating liquids within a total uncertainty of 5%, samples have been subjected to a Karl Fischer titration test applied to oils. The results have been compared to the limits given in the IEC and VDE standards.

In this paper, E represents a value for the ester liquid and M a value for the mineral oil. Thus M/10/20 for example signifies a mixture of mineral oil with 10% ester liquid and a water content of 20 ppm.

5.1 Determination of the relative permittivity and the dissipation factor of the mixed liquids

A liquid mixture can be considered as two different dielectric layers, where the indices 1 and 2 represent the mineral oil and the ester liquid respectively, the thickness d1 and d2 of each depends on the percentage of each liquid. The equivalent relative permittivity of the mixture having a thickness d is given by:

\[ \varepsilon_{eq} = \frac{\varepsilon_1 d_1}{\varepsilon_1 d_1 + \varepsilon_2 d_2} \]

The equivalent dissipation factor of the mixed insulating liquids, treated as two different layers, is:

\[ \tan \delta_e = \frac{\varepsilon_1 d_1 \tan \delta_1 + \varepsilon_2 d_2 \tan \delta_2}{\varepsilon_1 d_1 + \varepsilon_2 d_2} \]

Table 2 summarises the calculated values of the relative permittivity and the dissipation factor of the liquid mixture according to (3) and (4) respectively. The relative permittivity \( \varepsilon_1 \) varies between 2.1 and 2.3 and \( \varepsilon_2 \) between 3.2 and 3.4 while the dissipation factor \( \tan \delta_e \) lies between \( 10^{-4} \) and \( 10^{-3} \) and \( \tan \delta_e \) between \( 3 \times 10^{-4} \) and \( 30 \times 10^{-4} \).

<table>
<thead>
<tr>
<th>Ester liquid amount (%)</th>
<th>0</th>
<th>10</th>
<th>20</th>
<th>50</th>
<th>100</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \varepsilon )</td>
<td>2.2</td>
<td>±0.1</td>
<td>2.275</td>
<td>±0.1</td>
<td>2.355</td>
</tr>
<tr>
<td>( \tan \delta_e )</td>
<td>5.5</td>
<td>±4.5</td>
<td>12.3</td>
<td>±10.7</td>
<td>19.7</td>
</tr>
</tbody>
</table>
The measurements of permittivity and dissipation factor have been carried out at temperatures of 25, 60 and 90°C and at various water contents with a Schering-Bridge having a sensitivity $5 \times 10^{-7}$ at 50 Hz. There is an increase in $\varepsilon_r$ with increasing proportion of ester liquid in a mineral oil/ester liquid mixture as it is already shown in Table 2. There is good agreement between the experimental and theoretical results for all the liquid mixtures studied. The temperature dependency of the dissipation factor of the aged and unaged specimens of mineral oil and liquid mixtures is shown in Figure 2, in which the values of tan $\delta$ are the average for all water contents at a given temperature. It can be seen, that the tan $\delta$ values at all temperatures are in reasonable agreement with those obtained theoretically in Table 2.

An accelerated aging leads to an increase in the tan $\delta$ values, but they are well below the limiting value of 1 required by VDE [14] for aged oils. Furthermore it can be seen, that the higher the proportion of ester liquid, the lower the aging rate. A large amount of ester liquid mixed with mineral oil seems to have a beneficial effect on the aging of the mineral oil mixture.

![Fig. 2: Dissipation factor of new and aged ester liquid / mineral oil –mixtures](image)

5.2 Breakdown Voltage ($U_B$)

The determination of the breakdown voltage has been carried out with a test set up according to Figure 3.

![Fig.3: Measuring circuit for breakdown tests.](image)
The temperature dependency of the breakdown voltages for mineral oil and liquid mixtures is shown in Figures 4, 5 and 6. The breakdown voltages shown are the average values for all water contents at a given temperature. The results for the aged liquid mixtures are sometimes higher than those of unaged liquids particularly for high proportions of ester liquid, for example 50% ester liquid. Ester leads in all cases to a higher breakdown voltage at lower temperatures. At higher temperatures the influence of ester on the breakdown voltage is negligible.

The breakdown voltage of the mixtures is less temperature-dependent than those of the pure mineral oil. The reason is the difference in the water saturation limit. All the breakdown voltages for new and aged liquid mixtures are greater than the limit of 50 kV at 20°C required by VDE [12] for unused oils.

The ester liquid is beneficial for aged mineral oil as well as when they are both aged together. This aspect can be very helpful when retrofilling service-aged mineral oil filled transformers with liquid mixtures.
5.3 Viscosity
The viscosity of an insulating liquid is important for heat dissipation and the impregnation process. It is a basic parameter in design calculations for heat transfer by either natural convection in smaller self-cooled transformers or forced convection with pumps in larger units [1]. A low value of viscosity and a good specific heat capacity are needed to achieve a high heat transfer capability. However, high values of viscosity, such as that of ester liquids, have, compared to mineral oils, the advantage of quickly reaching the expected service temperature during a cold-restart of the transformer [11]. As viscosity decreases with the temperature, the viscosity of ester liquid at service temperature is relatively close to that of mineral oil. Generally, the viscosity of a mixture increases with increasing molecular size and molecular weight of the constituent compounds [1]. The viscosity of mineral oil is very low compared to that of ester liquid. By mixing the two liquids, an increase in the viscosity due to the proportion of ester liquid is expected. Figure 7 shows the measured viscosity of the mixtures as well as the pure liquids at two different temperatures. As expected, the higher the proportion of ester liquid, the higher the viscosity of the mixture.

5.4 Gassing behaviour of the insulating liquids at thermal stress
Insulation damages during operation are normally accompanied by gas production (and can cause PD). The gas can either be dissolved in the liquid or it can be free. The dissolved gases are detected with Dissolved Gas Analysis (DGA) methods [15, 16]. The
undissolved gases, which are generally produced by large faults in a short time or by a small faults long time after gas saturation in the liquid, ascend on their way up into the Buchholz-relay [16]. The dissolved and undissolved gases have been used since many years for the determination of the insulation condition (over-voltage, over-current, temperature of the hottest spot, etc.).

The thermal stress of the liquid specimen have been simulated in a set up according to Figure 8. It consists of a Borosilicat-vessel and a PTFE cover with clamps to fix the heating wire. The heating wire is made of Konstantan. The temperature around the heating wire is assumed to be constant, it was measured with a NiCrNi-temperature sensor. The heating current is delivered via a power current transformer. With an amperemeter connected to the primary and secondary windings the temperature at the wire can be selected. The funnel-shaped PTFE device over the heating wire allows the generated gases to ascend directly to a burette, from where they can be measured. This test set up allows a local controlled heating of the liquid up to >1000 °C.

The calculation of the local temperature of the liquid surrounding the heating wire is obtained using a thermodynamic approach. In the area of the heating wire the temperature is assumed to be constant, the radiated heating energy is therefore:

\[
Q_{ab} = \alpha_{Str} \cdot A_{heating} \left[ \frac{T_2}{100} \right] \left[ \frac{T_1}{100} \right]
\]

(5)

\[Q_{zu} = P = I^2 \cdot R\]

(6)

\[Q_{zu} = I^2 \cdot \frac{\rho \cdot l_{H}}{d_{H}^2 \cdot \pi / 4}\]

(7)

![Image](image)

Where:
- \(Q_{ab}\) Radiated heat power
- \(Q_{zu}\) Supplied thermal output power
- \(\rho\) specific resistivity
- \(\alpha_{Str}\) Radiation coefficient (3...5.77 W/m²K⁴)
- \(d_0\) Area of constant temperature (\(\phi \approx 10-20\) mm)
- \(A_{heating}\) Heating wire surface in the isolating liquid
- \(T_1\) Surrounding temperature (\(T \approx 298\) K)
- \(T_2\) Temperature in the region \(d_0\)
- \(d_{H}\) Diameter of the heating wire
- \(l_{H}\) Length of the heating wire

With the assumption of a constant temperature around the heating wire (10mm \(\leq d_0 \leq 20\)mm) and a radiation coefficient \(\alpha_{Str} = 3...5.77\) W/m²K⁴, the results of the calculation of the heating temperature (around the wire) are depicted by Figure 9. This Figure shows the results of the investigations performed for a maximal current of 50 A on the assumption of a homogeneous heating area \(l_{H}\) of approximately 15 mm around the heating wire with maximal temperatures of over 800°C. Accompanying measurements acknowledged the calculation.

For the mineral oil the first noticeable undissolved gas initiation appears at a temperature ranging from 250 to 300°C while for the ester liquid a temperature of 350 to 400°C is required. Through a small increase in the temperature, an exponential gassing tendency for the investigated insulating liquids is observed. At a given temperature, the total amount of undissolved gas generated by mineral oil is much higher, than that of ester liquid as previously investigated [15]. As an example, the behaviour of the mixture with 20% ester liquid is depicted in Figure 9.
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Figure 8: Test vessel and set up for local heating of the insulating liquid.

Fig. 9: Calculated temperature $T_2$ at the heating wire versus the heating current for different emission coefficient and different zones with constant temperature around the heating wire ($\alpha_{\text{str.}} = 3...5.77$ W/m$^2$K$^4$).
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At a given temperature, mineral oil generates a much higher amount of undissolved gas than ester liquid and the mixture of mineral oil with 20% of ester liquid. It has to be noticed, that the solubility of gases such as hydrogen is low in insulating liquids and that gas bubbles can be formed in the liquid (in the vicinity of discharges or local heating). Gas bubbles are weak dielectrics and their presence can intensify the discharge – corona- or initiate further breakdowns in highly stressed regions nearby. The higher the gassing tendency, the higher the risk of breakdown inception in the insulating liquid. Adding ester liquid to mineral oil helps decreasing the gassing tendency under local thermal stresses.

6. Conclusions

Ester liquids have good electric, dielectric, thermal, aging and environmental characteristics. Furthermore they have - in comparison to mineral oil - much lower inflammability and thus much lower fire risk. As moisture is 'enemy number one' for transformer insulations, the high saturation limit of ester liquid, almost more than 50 times higher than that of mineral oil, reduces the moisture content in the solid insulation due to water diffusion into the liquid, and as a result, the dielectric properties of the mixed liquids are changed slightly. If necessary, excess moisture can be removed from the fluid using standard techniques. Altogether, if the transformer usually operates at very low temperatures, the application of the mineral oil and ester liquid mixtures offers increased insulation reliability. The dielectric strength at low temperatures is higher, than that of pure mineral oil. Therefore, the lower risk of breakdown of the insulating liquid at low temperatures reduces the probability of a transformer malfunction. Ester liquids and mixtures of ester and mineral oil have in comparison to mineral oil a lot of advantages as insulating and cooling medium in transformers.

The electrical and physical properties of the investigated mixed liquids are not inferior to those of typical transformer oils, particularly for mixtures with 20% ester content or less. For the mixture with 50% of ester liquid the density and the cinematic viscosity exceeds the limiting values recommended by the standards for pure mineral transformer oil.

7. References

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[14] IEC 60156 (VDE 0370 Teil 5) "Insulating liquids: Determination of the breakdown voltage at power frequency", 1995