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Dielectric properties enhancement of vegetal transformer oil with TiO_2, CuO and ZnO nanoparticles

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Abstract. Nanofluids based on vegetal oils remain almost unexplored, especially when prepared with non-ferric oxide nanoparticles. This work has analysed the effect of three different nanoparticles on properties of a commercial vegetal oil. The first aim was to evaluate if there is an optimal concentration for dielectric improvement, and if the effect of different nanoparticles is similar. Secondly, other key properties were considered. The evaluated dielectric properties have been AC breakdown voltage, relative permittivity, volumetric resistivity and dissipation factor. The results are quite promising because dielectric properties have shown a considerable improvement. On the other hand, thermal conductivity or acidity test results, although adverse, were not definitive to refuse to continue this research.

Key words

Breakdown voltage, dissipation factor, vegetal oil, nanoparticles, nanofluids, transformers.

1. Introduction

Ester dielectric oils provide different advantages in comparison with mineral oil, such as higher flash point, higher thermal conductivity and biodegradability [1], [2] or better protection against ageing of cellulosic components [3], [4].

However, these alternative fluids must face yet different challenges to replace mineral oil. The major challenges include price, homogeneity of the product, as well as comparatively worst behaviour as coolant [5], [6], due to their higher viscosity, which hinder its acceptance by transformers manufacturers. This last issue might be overcame through thermal and dielectric properties enhancement of vegetal oils using nanoparticles.

Several investigations, mainly developed with traditional mineral oils as base fluid, support this statement, for both thermal [7]–[12] and dielectric properties [9], [10], [13]–[15], although there are also discrepancies [10].

Taking as examples those carried out with vegetal dielectric oils, according to corresponding standards, it has been proved enhancements respecting base fluid of breakdown voltages, and same occurs with their coolant capacity. These include maximum increases between 8 and 65%, under different kind of stresses, as AC or lightning impulse voltages [11], [13], [16]–[19]. Regarding thermal capacities, an increase until 45% in warming velocity of samples was noticed [11], what was understood as an enhancement on heat transmission through the fluid.

These results, although encouraging, are scarce and completely based on nanofluids with iron oxide nanoparticles. Moreover, references also showed the effect of preparation parameters as concentration, size, moisture or temperature on the improvement of properties [7]–[12], [14]–[18][20]

Therefore, it is necessary to study the behaviour of a wide range of nanofluids based on other nanoparticles, which possesses different properties, to evaluate if their effect on vegetal dielectric oils is comparable to iron oxides based nanofluids, and overall with those nanofluids prepared with mineral oils. At the same time, the study of different nanofluids requires evaluate all the variables that modify fluid final properties.

This article, as a first attempt in this way, addresses dielectric characterization of three different nanofluids, containing zinc, copper and titanium oxide commercial nanoparticles, added in six different concentrations in natural-ester transformer oil.

2. Nanofluids preparation

Nanofluids used in this paper were prepared in 5 different weight ratios from 0.01 to 0.2 % (Figure 1), plus a sample of base fluid that suffers same preparation steps than others to compare the results. These concentrations were chosen to identify the optimal one, as it is often between 0.01 and 0.05% mass fraction [11], [15].

Each calculated amount of oxide nanoparticles was added to 700 ml of base fluid, contained in 1 l Erlenmeyer flasks, and summited to 40 minutes of magnetic stirring at rated temperature with the flask open. Once dispersions were homogeneous, every sample was split into three 250 ml

sample bottles, that kept closed during 12 hours in an ultrasonic bath. Latterly, samples remained static for 24 extra hours, to disperse bubbles created during preparation, as their presence is detrimental for parameters to be measured. These steps are the most frequent when preparing nanofluids, as can be seen in the references mentioned in this article [7], [8], [11], [15]–[19][20] With samples ready, it started with dielectric tests.
It must be pointed that, as an initial approach, in this case, no additional treatments, as drying of samples [9], [11], [14], [17]–[19] or coating [7]–[11], [15]–[19] and washing [18], [19] of particles, were performed. In the same way, an optimum dispersion process was not pursued.

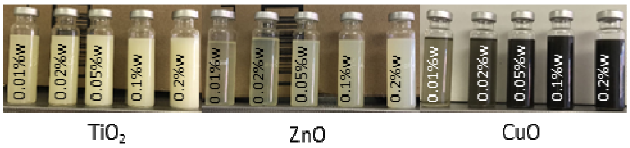

Fig. 1. Samples of studied nanofluids.

3. Characterisation tests

This research, as it was said before, just seek to reflect nanofluids first stage, does not pretend to consider time evolution of nanofluid and their properties, to allow conclusions just about nanoparticles effects due to their presence, type and concentration. The main reason that justified this decision is that by modifying and studying these parameters enough information may be obtained to reject the suitability of these particles, if it is the case. If not, other variables will be involved subsequently in future researching.
For this, tests for every combination of base fluid and particle were also carried out during the same day, with carefully homogenized samples, to ensure equal conditions. Dielectric, thermal and physic properties that conditionate suitability were investigated.
This work has carried out tests of AC breakdown voltage (IEC 60156), to define dielectric strength of prepared nanofluids, and measurements of relative permittivity, volumetric resistivity, and dissipation factor (IEC 60247), as indicators of their dielectric quality. Oil testers Baur DPA 75 C and Baur DTL 2a were used. Related with these, moisture content measurements were also done, with coulometer Metrohm 899 by Karl-Fischer method (IEC 60814), to certificate similar conditions of samples and to enable the comprehension of dielectric results.
Other thermal and physical properties as viscosity, density and thermal conductivity were measured, by HAAKE VT550, Mettler Toledo DM40 and Gunt WL422 respectively. The cooling performance of a fluid depends in part on these.
In the same way, acidity could conditionate the possible application of an insulating fluid, in case its level were harmful for other components. Titrator Metrohm 848 helped controlling it, according to ASTM D664 – 11a.
Additionally, to get some idea about the problem of stability of prepared fluids, samples of everyone, around 20 ml, were taken and stored in sealed vials. They were maintained static for days to assess their evolution with time by sight, registered by photographs.

4. Results

A. Dielectric strength.

Resistance to electric failures of transformer oil depends mainly on this parameter, represented by breakdown voltages, so a substantial increase of this would be definite to support vegetal nanofluids application in transformers.
Figure 2. shows breakdown voltages obtained in this investigation for different concentrations of nanoparticles in comparison to base fluid (0%w). In same conditions, presence of nanoparticles improves this parameter, although the optimal concentration to obtain the maximum value is different for the three studied nanofluids. These maximum increases are around 44, 32 and 11% for TiO_2, ZnO and CuO nanofluids, at 0.02, 0.02 and 0.01 % weight fraction concentrations, respectively. Both optimal increases and concentrations match with those found in references. In the case of CuO nanoparticles, the breakdown voltage increases firstly, but finally it decreases compared with base fluid. This behaviour is opposite to the other nanofluids, but every of them are above 35 kV, the minimum stablished in standards for natural esters (IEC 62770).

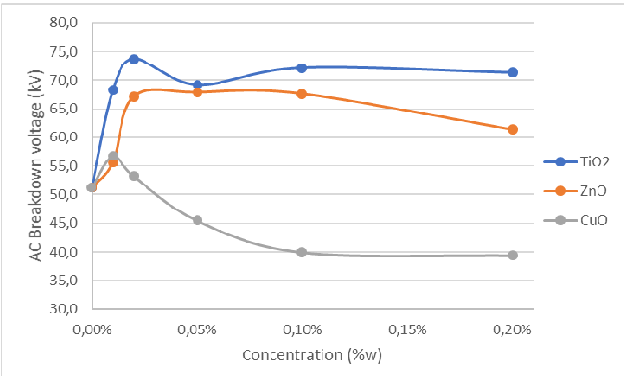

Fig. 2. AC breakdown voltage evolution with concentration.

A spread theory that explains this behavior is that nanoparticles creates potential traps on their surface under electrical fields, as those generated during dielectric tests. These traps, according to this theory, would be capable of act as sinks for free electrons, which are the main cause of the formation of streamers, which are the mechanism of the dielectric breakdown [10], [11], [13]–[17]

B. Dielectric relaxation.

This includes yet mentioned relative permittivity (ε), volumetric resistivity (R) and dissipation factor (tan δ). These are interdependent, as dissipation factor represents conductivity and polarization losses [16]. All them were measured at rated frequency.
Permittivity remained constant in every measured sample, in 2.8, so polarization losses had no effect on dissipation factor variation, as in references [9], [16]. This is thus ascribed to changes in resistivity. In Figures 3 to 5. both are plotted, showing opposite tendencies, as expected [16]. It is also clear the beneficial effect of nanoparticle presence on these parameters, as dissipation factor decreases and resistance grows, becoming better

insulators. Again, CuO particles show the worst behaviour.

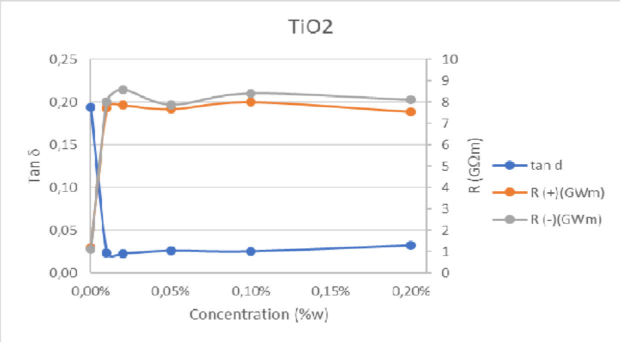

Fig. 3. Dielectric relaxation of tested TiO_2 nanofluids.

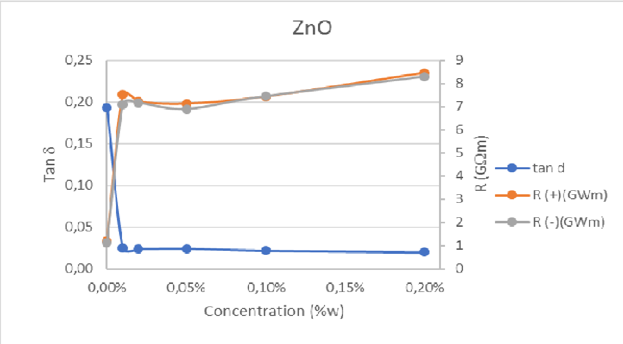

Fig. 4. Dielectric relaxation of tested ZnO nanofluids.

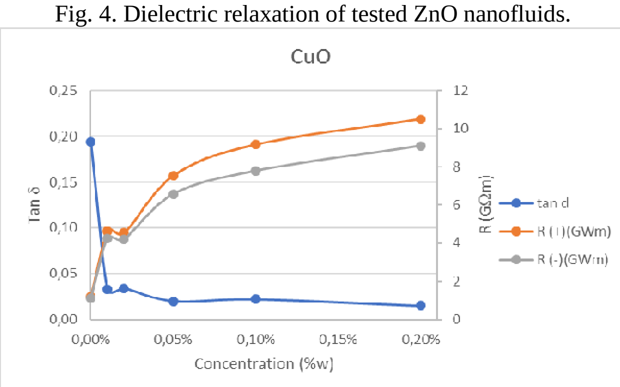

Fig. 5. Dielectric relaxation of tested CuO nanofluids.

C. *Thermal properties*

The equipment available for thermal conductivity measurements in this research was one for educational proposal, based on directly measured thermal gradients, more inaccurate than other alternatives usually found in references [9], [12], so these results must be treated with caution.

In Figures 6, 7 and 8., plots seem very changing, without a clear tendency and always under theoretical capacities of base fluid. Although, it seems that there is a change on evolution of thermal conductivity, especially considering the results at higher temperatures, where the accurate of equipment improves.

The conductivity of dielectric oil tends to decrease as temperature grows [10], contrary to water, what can be seen in manufacturers base-fluid line (0%), but here experimental results behave reversely, as it was noticed in other references [9].

Neither the optimal concentration nor which particle is best for thermal conduction can be inferred from these results. However, once again copper oxides nanofluids always perform worse than their alternative peers (an example is shown in Figure 9.), and the variability in results between different concentrations could point to the existence of this optimal concentration, as in other references [9], [10].

D. *Physic properties.*

Both density and viscosity have little variations respecting base fluid, due to low concentrations of nanoparticles in suspension. At rated temperature, maximum variation in viscosity was under 5 %, while density remained constant.

E. *Moisture.*

As can be seen in Figure 10., moisture content in all samples are in the same order of magnitude, but not equal. A slightly tendency to grow is observed in all the nanofluid combinations with nanoparticle concentration. It is also noticed that fluid base moisture absorption is lower.

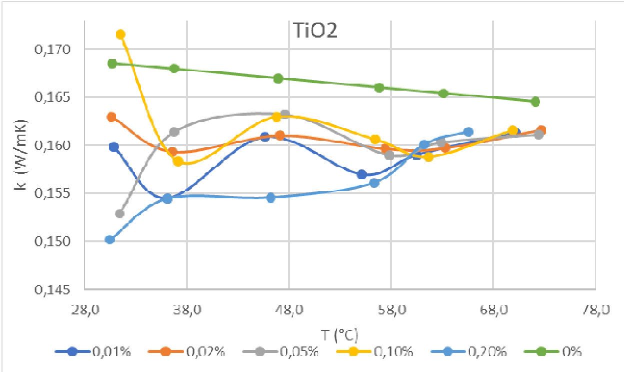

Fig. 6. Thermal conductivity of tested TiO_2 nanofluids.

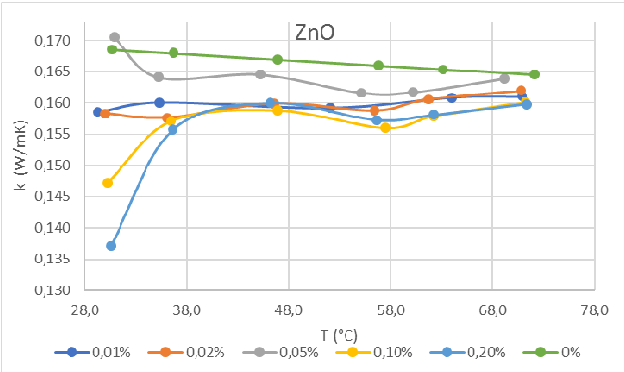

Fig. 7. Thermal conductivity of tested ZnO nanofluids.

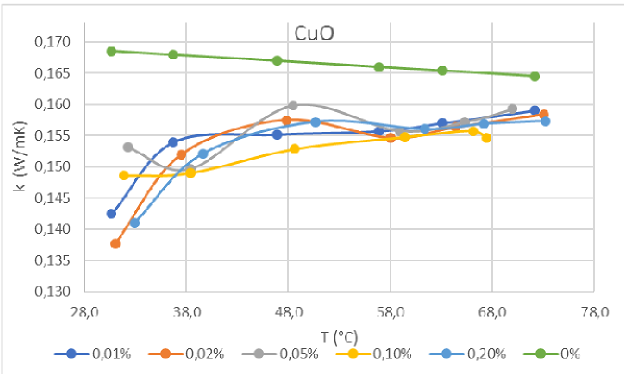

Fig. 8. Thermal conductivity of tested CuO nanofluids.

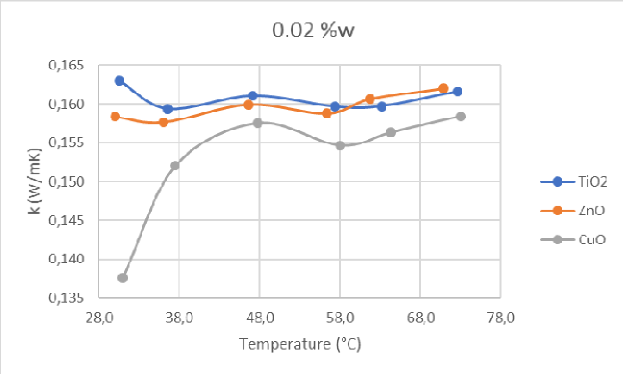

Fig. 9. Thermal conductivity of 0.02% weight fraction fluids.

So, presence of nanoparticles seems to increase water sorption, probably in their surface [14], [15] yet this is not translated to a worsening of breakdown voltages or dissipation factor of base fluid, which actually were observed in other references [14], [18], [20]. Nevertheless, it is possible that the decrease in breakdown voltage from the optimum concentration, at least in part, was due to higher moisture content in these samples. To clarify this situation, moisture controlled dielectric tests will be needed.

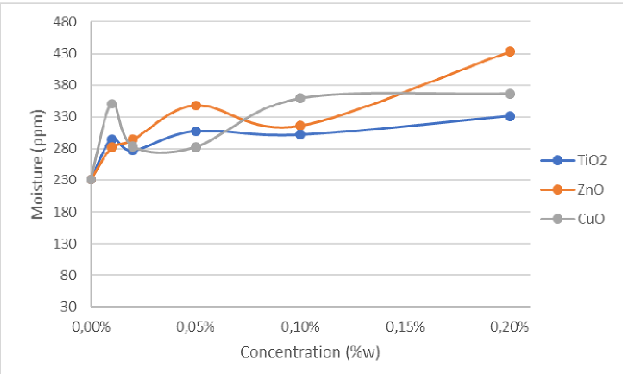

Fig. 10. Moisture content on prepared samples.

F. Acidity.

According to IEC 62770, acidity of a new natural ester dielectric oil must be under 0.06 mg KOH/g oil. As can be seen in Figure 11., all the prepared nanofluids exceed this limit, which is 6 times higher to that established for mineral dielectric oils (IEC 60296). With this values it is pretended to protect cellulosic dielectrics in transformers from degradation, but many references have pointed that in fact, the presence of nanoparticles in dielectric fluids hinders this phenomenon [3], [4], so it is not clear that this maximum value of acidity is suitable for dielectric nanofluids, and these nanofluids cannot be rejected because this reason.

Overall, although unstable, it seems that acidity grows together with nanoparticle concentration.

G. Dispersion stability with time.

Although it was not an objective of this research, samples of nanofluids were taken, to observe how long their last would be. Their irregular evolution is collected in Figure 12. While copper oxide nanofluids started sedimentation faster, as were almost sedimented in 5 days, and completely in another 7 extra days, titanium and zinc oxides nanofluids maintained their dispersion, with signs of sedimentation, for more days. Some of them kept stable after 4 weeks, especially those less concentrated, which are at the same time the most interesting due to their dielectric properties.

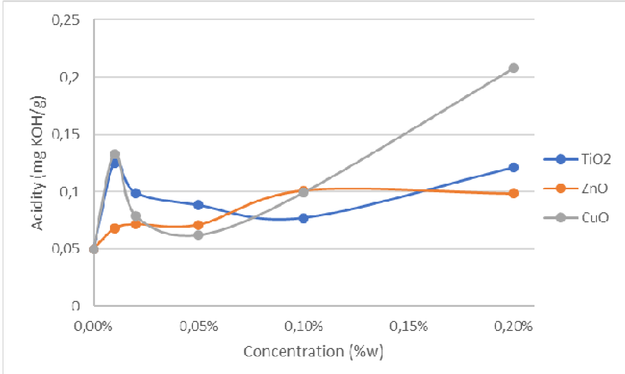

Fig. 11. Measured acidity in tested samples.

A plausible explanation for CuO nanofluids performance comes from stability tests. As these particles sediment earlier, probably they suffer more aggregation, that increases nanoparticles size and reduces both available surface for electron trap and probability of encounters between particles, harming dielectric strength and thermal conductivity.

It is noticeable that some concentrated samples are finally more like base fluid than the other, that maintain part of the colour of particles, as if particles bring each other down easily in these cases.

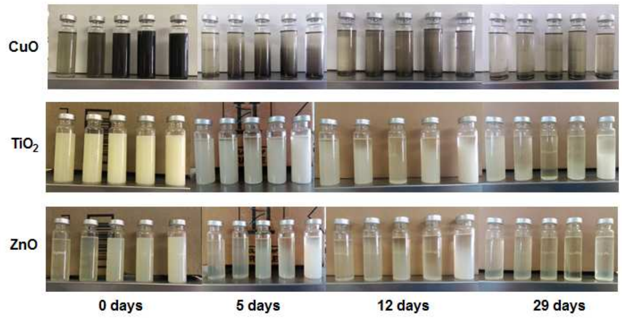

Fig. 12. Samples appearance 0, 5, 12 and 29 days after tests.

5. Conclusions

The addition of different nanoparticles to a commercial vegetal oil has shown to be positive under the point of view of the dielectric properties. An increase in resistivity and a decrease in the loss factor have been achieved for the three studied nanoparticles, improving insulating capacity of the dielectric oil. On the contrary, presence of nanoparticles increases acidity, and decreases thermal conductivity, but these are not decisive by themselves to reject these nanofluids. Therefore, they become suitable alternative to iron oxides, but this could not be applicable for CuO nanofluids, as their performance in tests is always the worst.

Future research should verify these conclusions, introduce extra parameters and variables not treated here, and study if the improvement of the dielectric properties is kept over time. This analysis requires an exhaustive study of the nanofluids stability.

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