

ENC-2022-0724

Experimental Evaluation on the effect of magnetite in aerogel form on the dielectric breakdown strength and thermal conductivity of a synthetic isolating oil

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Abstract. Nowadays, the recent trend of miniaturizing power electronics systems led to higher power dissipation per unit area, demanding an enhancement of thermal properties of isolating oils. This study proposes to improve the thermal conductivity (while having admissible dielectric breakdown strength) of widely used commercial synthetic oil, by adding magnetite in aerogel form. Mass fractions of nanoparticles and aerogel of 0.01% and 0.05% were investigated experimentally, and the results were compared with those of the base fluid. A FOX 50 Thermal Conductivity Meter was used to assess thermal conductivity and a HMRDT-100 dielectric breakdown testing equipment measured the breakdown voltage through the short-time method with controlled fluid temperature. The results indicated an increase of thermal conductivity and electric breakdown strength in the majority of the test samples when compared to the isolating oil. An increase of 22% of the electric breakdown strength and 1.5% of the thermal conductivity was observed for the 0.05 wt% Fe_3O_4 aerogel sample. Therefore, the addition of Fe_3O_4 aerogel to commercial synthetic isolating oil is a promising alternative to nanoparticles to enhance thermal conductivity and electric breakdown strength, due to their smaller particle size and consequently larger surface area which lead to better dispersion stability. Yet, higher mass fractions of magnetite aerogel in isolating oil must be investigated and other thermal properties should be assessed in order to establish it as a novel coolant for electronics.

Keywords: magnetite, aerogel, isolating oil, electric breakdown strength, thermal conductivity.

1. INTRODUCTION

Electronic components had their power density increased greatly in the last years, demanding an improvement on the existing cooling techniques to achieve a higher heat extraction capability. In this context, passive cooling systems using isolating oil play a significant role, since they are used to ensure proper thermal management of transformers and subsea frequency inverters (Militao *et al.*, 2021; Boe, 2010, 2011; Bakka *et al.*, 2001; Parente, 2015; Lendenmann *et al.*, 2019). Recent studies aimed to increase the performance of such fluids by enhancing its thermal properties, while maintaining its dielectric capability (Munaro, 2017; Krishnam *et al.*, 2016; Dalla Vecchia, 2019).

Dalla Vecchia (2019) dispersed weight fractions of 0.01%, 0.05%, 0.10%, 0.50%, 1.00 % and 2.00 % of Fe_3O_4 in MIDEL 7131 synthetic oil, and measured density, dynamic viscosity and thermal conductivity of the nano-oil and compared them to the base fluid thermal properties. An increase of 1.5% was obtained for the thermal conductivity, for the 2.0% wt concentration. It was found that the increase in thermal conductivity decreased with the temperature rise of the nano-oil. Therefore, the enhancement of thermal conductivity in the base fluid is more pronounced at lower temperatures.

Du *et al.* (2015) investigated the effect of dispersing Boron Nitride (BN) and Magnetite (Fe_3O_4) in mineral oil on the thermal conductivity and dielectric characteristics. Weight fractions of 0.05% and 0.1% of BN and Fe_3O_4 were investigated. An increase of the thermal conductivity of 0.4% was obtained for the Fe_3O_4 0.1% weight fraction. An increase in the AC Breakdown Strength of 55% and an enhancement of 42.8% in the DC Breakdown Strength was obtained for this same weight fraction.

Cavallini *et al.* (2015) evaluated the influence of the dispersion of Fe_3O_4 , graphene oxide and SiO_2 in the dielectric withstand capability of mineral oil. It was found that a concentration of 0.2 g/l concentration of all nanoparticles would enhance breakdown voltage. However, this was not the case for divergent electric fields and equipment working with dc or ac+dc voltage waveforms. Nevertheless, a consensus among nanofluids' researchers is that dispersion stability is a problem, since it depletes with time, hindering the heat transfer and dielectric withstand capability (Mukherjee and Paria, 2013; Du *et al.*, 2015).

On the other hand, aerogel nanoparticles are expected to have greater dispersion stability due to their much smaller particle size. In this scenario, dispersing metallic oxide aerogel in dielectric oils might be a good alternative to increase the dielectric breakdown strength and enhance the thermal conductivity for long periods of time due to their dispersion stability. Therefore, this study proposes a stable nano-oil with considerable dielectric characteristics and enhanced thermal conductivity by dispersing Fe_3O_4 in synthetic dielectric oil. Weight fractions of 0.01% and 0.05% of aerogel nanoparticles and regular nanoparticles were dispersed, and the breakdown voltage and thermal conductivity were measured to determine if aerogel nanoparticle dispersion of promising metallic oxide material is indeed a better solution than its regular nanoparticle counterpart.

2. MATERIAL AND METHODS

With the objective of comparing the performance of nanofluids synthesized with nanoparticles or aerogels, various weight fractions of hBN and Fe_3O_4 were dispersed and analyzed. The nanofluid samples were obtained dispersing nanoparticles of Fe_3O_4 with concentrations of 0.01, 0.05 wt%, and the same concentrations were used with the aerogel particles. The thermal conductivity and electric breakdown strength were measured in the following temperatures: 10, 20, 30, 40, 50 and 60 °C.

The masses of Fe_3O_4 nanoparticles and aerogel were determined by multiplying the weight of base fluid and the desired weight fraction, calculated as shown by Eq. 1:

$$m_A = m_{bf}\phi \quad (1)$$

where m_{bf} is the mass of the base fluid, m_A is the mass of nanoparticles or aerogel and ϕ is the Fe_3O_4 nanoparticle or aerogel weight fraction wt%. After mixing, the nano-oil is weighted one more time to guarantee that the previously established fractions were attained.

2.1 Synthesis of Fe_3O_4 aerogels

The methodology reported by Zhang *et al.* (2014) was used as a basis for the synthesis of Fe_3O_4 aerogels. Briefly, $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (98% of purity) and $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$ (98% of purity) are dissolved in 10 mL of absolute ethanol (99.8% of purity) in a 2:1 ratio of Fe+3:Fe+2 under an Argon atmosphere. Into this resulting solution, 4 mL of propylene oxide (99% of purity) were dripped and poured into molds, obtaining the gels in 2 minutes. Subsequently, the gels were kept in an ethanol bath for 7 days to remove impurities and subjected to a supercritical CO_2 drying (Tousimis Autosamdri 931, USA) to form Fe_3O_4 aerogels.

2.2 Synthesis of Fe_3O_4 nanoparticles

The first step of the dispersion procedure is weighting the base fluid with a precision scale (*Sartorius model ED224S*) extend with an accuracy of 0.1 mg. Subsequently, the samples were homogenized using a high intensity ultrasonic processor, (*SONICS Vibra Cell VC 750W*) for one hour in a copper vessel. A critical aspect of the dispersion process is the need for a precise temperature control of the samples, which was ensured by using an ultra thermostatic bath to maintain the sample contained in a copper vessel at 5 °C. A schematic diagram of the dispersion process is shown in Fig. 1.

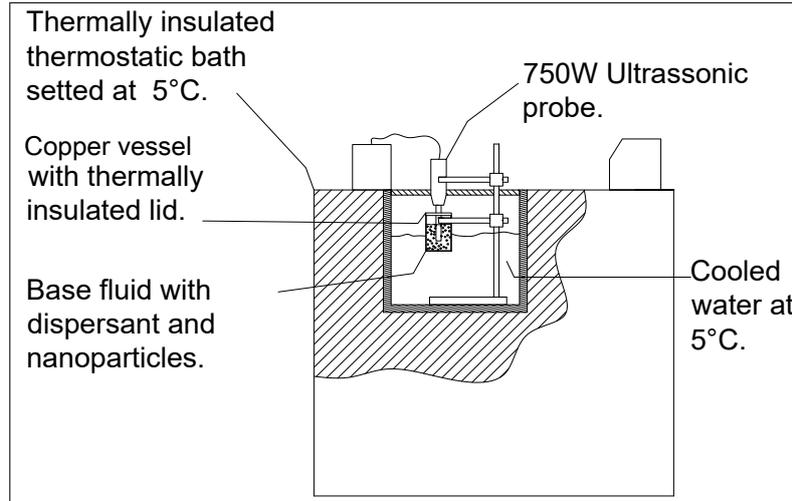


Figure 1. Schematic diagram of the dispersion procedure.

2.3 Characterization

The crystalline structure of the Fe_3O_4 aerogels was evaluated by X-ray diffraction using a XRD device, manufactured by Philips X-Pert, with $\text{Cu-K}\alpha$ radiation ($\lambda = 1.54 \text{ \AA}$) at a scan rate of $1^\circ/\text{min}$. The specific surface area of the samples was determined through nitrogen adsorption isotherms at 77 K by the Brunauer, Emmett and Teller (BET) technique, using the equipment *Quantachrome NOVA 1200e*, with degassing at 100°C for 2 h. The microstructure and morphology of the Fe_3O_4 aerogels were evaluated by transmission electron microscopy, model *JEOL-JEM-1011*, with a maximum acceleration voltage of 80 kV. The average particle size was determined by image analysis of TEM micrographs (ImageJ®) using at least three images.

To determine the thermal conductivity of the obtained samples, the *FOX-50* device manufactured by *TA Instruments* was used. The instrument has a thermal conductivity range of 0.1 to $10.0 \text{ W}/(\text{m}\cdot\text{K})$, the resolution of the temperature is $\pm 0.01^\circ\text{C}$, the accuracy is noted as $\pm 3\%$ and the reproducibility is $\pm 2\%$. The device was configured to measure in the specified temperatures $10, 20, 30, 40, 50, 60^\circ\text{C}$. The nano-oil samples were put inside a fluid cell, that was inserted between the plates of the instrument. A descriptive image of the *FOX-50* is shown in the Fig.2.

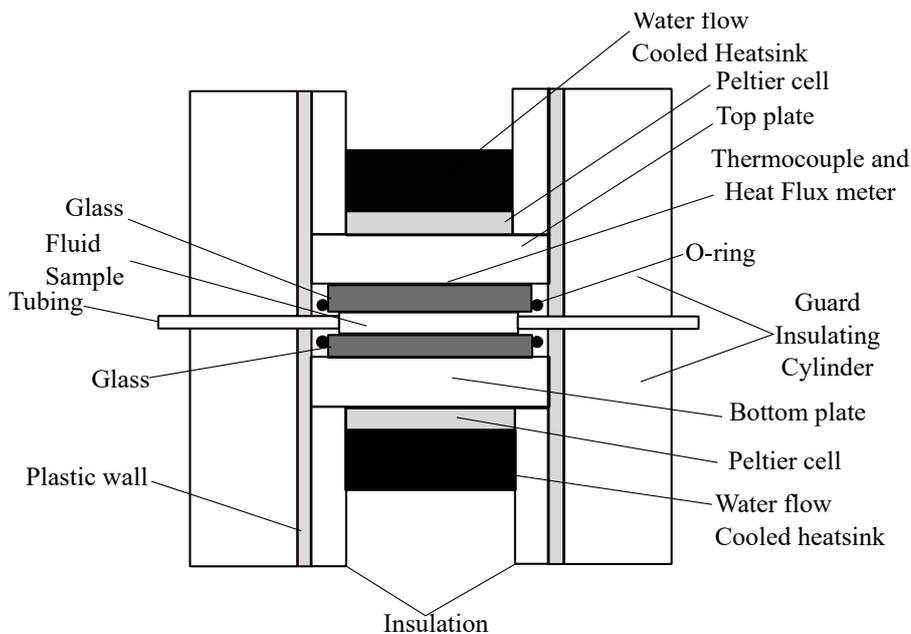


Figure 2. Device for thermal conductivity measurement, with fluid cell inserted.

In order to determine the thermal conductivity, the sample is placed between two thermal insulated metal plates that control the temperature within the specified range via Peltier Cells. Once the device attains steady state, the thermal conductivity is determined from Fourier's law by analyzing the heatflow and the temperatures via heat flux meters and thermocouples.

Dielectric strength tests (*Highmed, HMRDT-100*) of the samples were performed by submerging two electrodes in the nanofluid. Voltage is applied to the positive electrode and is increased in steps until the voltage is sufficient to break the dielectric strength of the fluid and pass current to the negative electrode. The electrodes are separated by a known distance d , with this information and the voltage required to break the dielectric strength of the sample, it is possible to calculate the Electrical Breakdown Strength (kV/mm). An accuracy of $\pm 1\%$ in this parameter is provided by the equipment. A representative image of the device is shown in Fig.3. All samples were analyzed in accordance with ASTM D 877, equivalent to NBR6869, which determines that, for the test, discoidal electrodes separated by 2.5 mm and a voltage rise ramp of 3 kV/s should be used, in the temperature range from 24 to 26 °C.

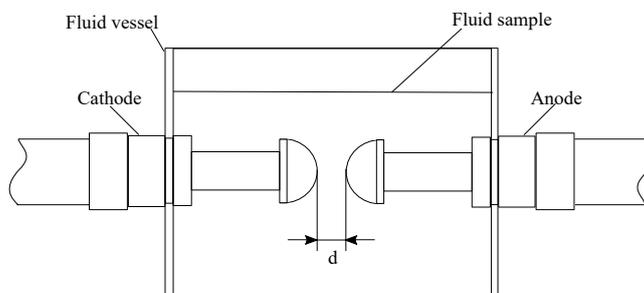


Figure 3. Frontal view of the apparatus used to measure the dielectric breakdown strength.

3. RESULTS AND DISCUSSION

Figure 4 shows the XRD diffractogram of the Fe_3O_4 aerogel sample. As expected, due to the absence of a thermal treatment step during the synthesis of the material, it presents a typical diffractogram of amorphous material, making it impossible to identify a crystalline structure of Fe_3O_4 in this analysis.

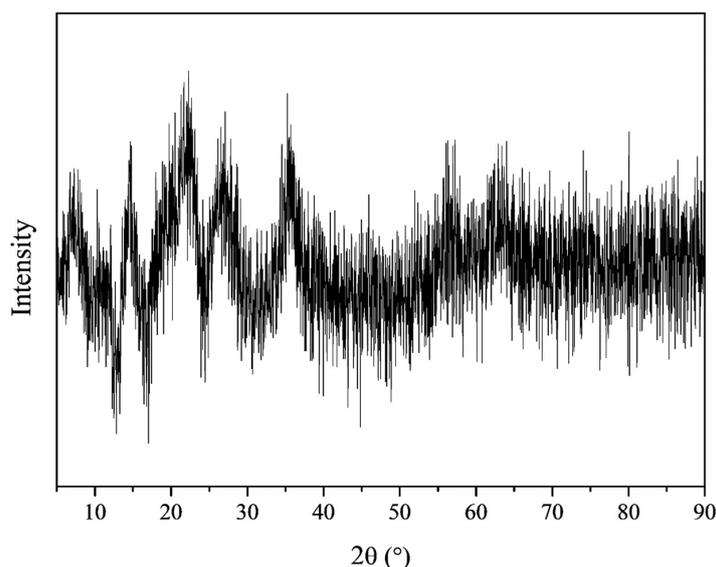


Figure 4. Fe_3O_4 aerogel sample XRD pattern

Figure 5 shows TEM micrographs in different magnitudes of the Fe_3O_4 aerogel sample. Fe_3O_4 aerogel exhibits a hierarchical morphological structure, with interconnected spherical nanoparticles forming a typical porous structure of aerogels, with average size of 4.0 ± 1.2 nm.

The specific surface area and mean particle size of synthesized and commercially available of Fe_3O_4 nanoparticles and aerogels is shown in Table 1. It is observed that due to the smaller particle size, favored by the structure of the aerogels, the surface area of the Fe_3O_4 aerogel samples is higher than the commercial sample, which presented a much larger particle size, causing a decrease in its surface area. With these data, it is expected that the aerogels present better dispersion stability in synthetic oil.

The Dielectric Breakdown Strength (kV/mm) and the Mean Breakdown Voltage (kV), for a total of five breakdowns and all considered test samples, is presented in Table 2, for a constant fluid temperature of 24°C. A higher standard deviation, σ , was observed for the nanoparticles sample with 0.05 wt% of Fe_3O_4 and the raw synthetic oil.

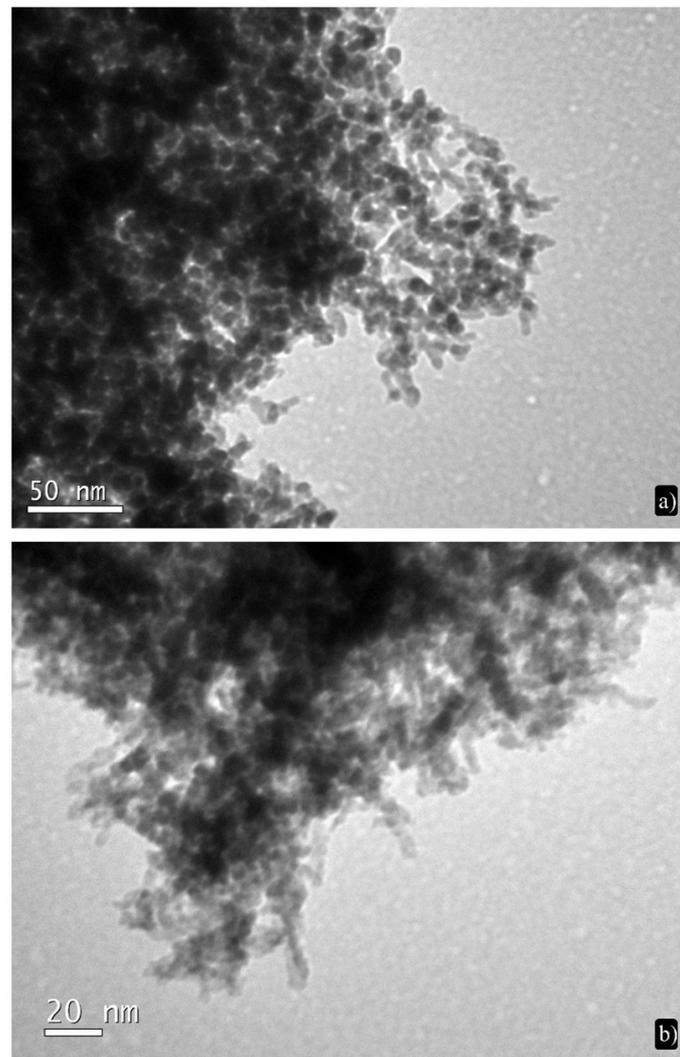


Figure 5. TEM images revealing the microstructure of the Fe_3O_4 aerogels in magnitudes of a) 50 nm and b) 20 nm.

Table 1. Mean particle size (Φ_M) obtained from TEM image analysis and BET specific surface area (SSA) of the Fe_3O_4 aerogels and Fe_3O_4 commercial.

Sample	$\Phi_M(\text{nm})$	BET(m^2/g)
Fe_3O_4 aerogels	4.0 ± 1.2	338
Fe_3O_4 nanoparticles	30	77

By analyzing the data, one can conclude that the addition of Fe_3O_4 nanoparticles to the base fluid deteriorated the dielectric breakdown strength, up to 25%, for the test sample with the higher mass fraction. On the other hand, when aerogel is regarded, there is a considerable increase at the property, of about 22% for the 0.05 wt% sample.

Next, the thermal conductivity values obtained for all test samples are presented in Figure 6, in comparison with bulk -MIDEL® 7131 data.

A clear similarity on the thermal conductivity behavior when the fluid temperature is increased is observed, as the curves are parallel to one another. Besides, the addition of Fe_3O_4 particles, regardless of their form, contributes to an elevation of overall thermal conductivity, for the whole temperature range. However, increasing mass fraction of nanoparticles did not increase significantly the property values, providing a maximum increase of 0.6%.

Furthermore, the aerogel-based synthetic oil showed a considerable higher thermal conductivity, for all temperatures, despite the mass fraction. A maximum increase of 1.5 % was observed for the 0.05 wt% Fe_3O_4 aerogel test sample.

Therefore, not only the addition of Fe_3O_4 in aerogel form increased dielectric breakdown strength of the isolating oil, but also enhanced the fluid thermal conductivity significantly, what could improve its use for electronic thermal management purposes. When compared to nanoparticles-enhanced Fe_3O_4 synthetic oil, its aerogel alternative shown to

Table 2. Dielectric Breakdown Strength for all test samples.

Addition	Sample	Mean Breakdown Voltage(V)	σ (kV)	Dielectric Breakdown Strength(kV/mm)
-	Synthetic Oil (MIDEL @7131)	40.8	7.5	16.3
Nanoparticles	SO+Fe ₃ O ₄ (0.01 wt%)	42.9	3.5	17.2
	SO+Fe ₃ O ₄ (0.05 wt%)	30.9	6.8	12.4
Aerogel	SO+Fe ₃ O ₄ (0.01 wt%)	49.6	2.2	19.8
	SO+Fe ₃ O ₄ (0.05 wt%)	45.8	2.4	18.3

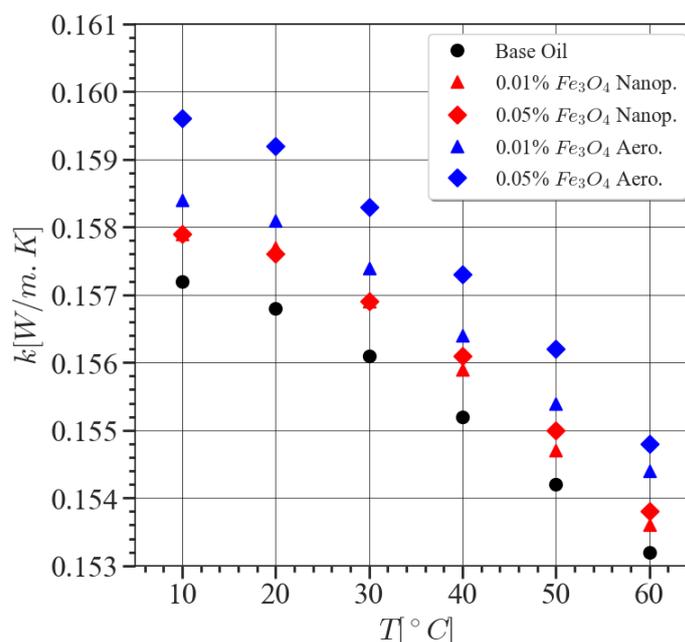


Figure 6. Thermal conductivity values for all test samples.

have a clear advantage, since the former provide smaller dielectric breakdown strength (inferior of the base fluid ones) and thermal conductivity values.

4. CONCLUSION

This study proposed to disperse nanoparticles and aerogel nanoparticles of Fe₃O₄ in synthetic isolating oil for different weight fractions, in order to observe the change in the thermal conductivity and dielectric breakdown strength and conclude if aerogel-based nano-oils are a more suitable option than nanoparticle-based nano-oils. Thermal conductivity measurements were taken at temperatures of 10, 20, 30, 40, 50, 60 ° C, and dielectric breakdown strength was measured for an ambient temperature of 25 ° C. An increase of the thermal conductivity was observed for all test samples (by comparing the base fluid to the nanofluid samples) either aerogel or nanoparticles-based, for the entire temperature range. Higher values were observed for the Fe₃O₄ aerogel samples, with a maximum 1.5% increase for the 0.05 wt% sample. As for the dielectric breakdown strength, again the Fe₃O₄ aerogel samples presented higher values, with a 22% increase in this property for the weight fraction of 0.05%.

Therefore, the Fe₃O₄ aerogel nano-oil is a superior choice when compared to the nanoparticle nano-oil and the base fluid, due to its superior thermal conductivity and dielectric breakdown strength, and its allegedly high dispersion stability. Future works could benefit from testing a wider range of weight fractions of Fe₃O₄ aerogel and nanoparticles in dielectric oil, and measuring dispersion stability through qualitative analysis and absorbance measurement. This would definitely establish Fe₃O₄ aerogel as a powerful enhancer of thermal and electrical properties of insulating oils used in passive cooling systems.

5. ACKNOWLEDGEMENTS

The authors would like to thank the Central Laboratory of Electronic Microscopy (LCME at Federal University of Santa Catarina) for TEM analysis and the multi-user facility LDRX at Federal University of Santa Catarina for the XRD analysis.

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