DIELECTRIC BEHAVIOR STUDY OF THERMOSETTING MATRICES LOADED

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Abstract:

The present study was prospective in nature and its purpose is twofold. It consists, on the one hand, to identify and characterize mineral fillers, secondly, to determine the dielectric properties of materials prepared using the epoxy resin mixed with local inorganic fillers , very rich in silica such that: sandy, pozzolan, cullet and a small calcium carbonate amount. This application allows us to use the matrix developed as an electrical insulator.

The results of our study show that the use of fillers further improves the dielectric properties of these matrices. After 32 days of conditioning, the values of capacitance and resistance obtained are very remarkable. Comparing our results with previous work with the addition of pure silica, we note that our composites have better properties at a low cost. The hygro-thermal influence is much less important for the loaded resin. Our composites may be subject to external environmental influences and they have a range of wide usage in dielectric materials.

Keywords: Matrix thermosetting, inorganic fillers, dielectric properties, resistivity, capacity

1. Introduction

The thermosetting matrix materials are most common in composite applications [1]. In this polymer type, the molecules are chemically linked together, forming a threedimensional network. Treatment can be achieved by the application of heat or by a chemical reaction [2].

Fillers or fillers are used in polymers for a variety of reasons, to reduce the cost, improve the treatment, control density, thermal conductivity, thermal expansion, electrical properties, magnetic properties, flame retardancy, and to improve the properties mechanical [3].

Each type of expense has different properties depending on the particle size, shape and surface chemistry [4, 5, 6]. The fillers used for most of the thermosetting resins are calcium carbonate, kaolin, alumina hydrate and [7]. The other filler commonly used include clay, carbon black, microspheres mica, and silica, glass, and glass favorites [8].

In composites based on epoxy resin, the dielectric properties are at first sight very remarkable, especially the resistivity is around 1015-1016 Ω • cm, but electrical equipment manufacturers as well as electronics especially appreciate the fact that they vary relatively little with the temperature and exposure time to moisture (as compared to other insulating materials).

The current measurements are used for a long time in the study of electrical insulators [9], [10]. In addition to the characteristic value of the resistivity, this technique allows to obtain information on the mode of conduction in materials.

More recently, Griseri et al. [11] observed behavior of current conduction with the electric field. This characteristic is representative of a limited current mechanism of space charge.

In general, the presence of fillers intrinsic, that is to say initially present in the material, generates straight load when electric field is applied. Charged species in the material migrating towards the electrode of opposite sign when voltage is applied. This has been demonstrated repeatedly in many studies conducted on the polyethylene.

And Hozumi [12] has detected straight load stable fields below 20 kV / mm, assigned to the dissociation of antioxidants. These same antioxidants increase the amount of space charges when their quantity increases and promote charge injection from the electrodes [13].

Ezoe also shown that in an epoxy resin straight load are derived from the acid groups of the material and their formation is favored by the presence of water in the material. The positive charges are then assigned to protons (H +) and negative charges species combined (R - COO-) [14].

Furthermore, Lee [15] is a decrease in the amount of straight load when species of smaller mass are removed from the material.

Finally, Li, who observed the straight load in a crosslinked polyethylene untreated no longer observed after degassing. The charges were granted in this case the reaction by-products [16].

2. Different materials used.

The matrix is an epoxy resin. It is obtained from two basic components; the stoichiometry is given by the supplier. These components are resin and hardener.

Mineral fillers used are respectively the sand, cullet and pozzolana with the CaCO3 addition. These loads are used because their high silica content. The loading rate of the composite is described in the next section.

2.1. Résine époxyde.

In our work, we used the MEDAPOXY STR of the company Granitex, They are known for their high performance and best qualities.

2.2. Mineral fillers

Mineral fillers were selected by their contribution to the silica content is important and their availability as raw materials such as natural or industrial waste, are highly sought as powders in various fields of science, on the one hand to improve certain characteristics or other share in order to reduce the cost of materials sought. In this study, the fillers used are calcium carbonate, sand, waste glass (cullet) and pozzolana. These powders are milled in a ball mill and sieved so as to use only the grain size less than 100 microns.

3. Geometric shape of the inorganic filler:

The particles can have different shapes: spherical, chipped, angular geometric shapes ... these depend on the nature of the charges and their world to obtain. Photograph taken with an electron microscope (Figure 1) shows the morphology the used fillers.

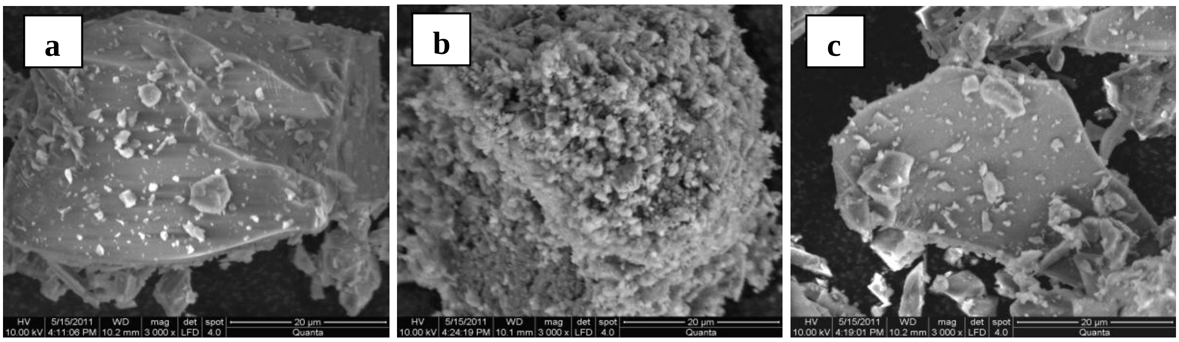

Figure 1 - SEM images (at 20µm) of grains:

- (a) Sand, - (b) Cullet - (b) Pozzolana

3.1. Loads size

It plays a role in the mechanical and aesthetic properties of composite resins and is the basis of the different those classifications.

The size and the shape will be developed in the next section on the synthesis of composites; in fact, the most commonly accepted classifications are based on the shape and size of the particles.

4. Synthesis of composite materials

The synthesis of the composite is made by mixing the epoxy resin and various fillers used. Formulations (Table I) were chosen according to the components and their sizes, as well as the charges geometry and their distributions size.

4.1. Material Preparation

The manufacturing protocol is identical to that industrial used, is described by the supplier Granitex. To observe the effects of interfaces epoxy / fillers, and spiked samples are made. In addition, these compositions are prepared with the addition of trihdroxysilane to assess the impact of surface treatment.

The day before the molding, the amount of charge required for the test loaded is weighed and placed in an oven at 60 ° C to dry. Two hours before the resin and hardener are weighed in the proportions indicated in Table I and are placed in an oven at 60 ° C to precondition the material. The resin is mixed with the hardener 15 minutes in a mixer preheated to 60 ° C under a low vacuum for degassing of the material and the removal of bubbles formed during the mixing phase.

The load is then incorporated into the mixture under the conditions indicated in Table I

Table 1 - Formulation of composites tested

Components		Size	Formulations		
			MSC	**MPC**	**MCa**
			Rate %		
Matrix	Resin		38	38	38
	Hardener		20	20	20
	Trimethoxyproyl	--	2	2	2
loads	Sand	<100µm	35	--	--
	Pozzolana	<100µm	--	35	--

Cullet	<60µm	--	--	40
$CaCO_3$	<20µm	5	5	--

4.2. Distribution and orientation of the fillers in matrix.

Distributions and orientations of loads in composite dimension strongly influence the final structure of the network samples. Mastery and understanding of the anisotropy and heterogeneity are crucial for improving the properties for this composites type. These characteristics are at the origin of instabilities geometric and tensions in parts. Changes in distribution and orientation may also cause surface deformation and thus have an impact on the final surfaces quality of the samples [17].

4.3. Measuring device

Resistance measurements are performed in the climatic chamber of 05 ° C to 150 ° C and 60% RH, under voltage of 1V and frequency of 1kHz and 10kHz, provided with a device developed in our laboratory (dielectric spectrometer at low voltage (THURLBY THANDAR INSTRUMENTS LCR400 TTi) is shown in Figure 2, it can be used over a wide frequency band including the frequency of transmission and distribution (1/10kHz).

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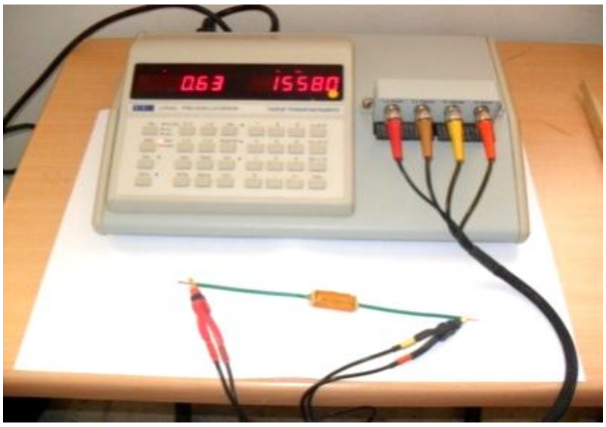

Figure 2 - low voltage dielectric spectrometer THURLBY THANDAR INSTRUMENTS (TTi LCR400)

4.4. Geometry samples

Represents in figure3, has a coating of copper inserts which act as electrodes. Their profile is used to generate a uniform field in the space between electrodes on a diameter of 0.5 mm. The external sample geometry is designed to prevent the breakdown. Indeed, the creepage is about 5 mm for an inter-electrode distance of 1 mm.

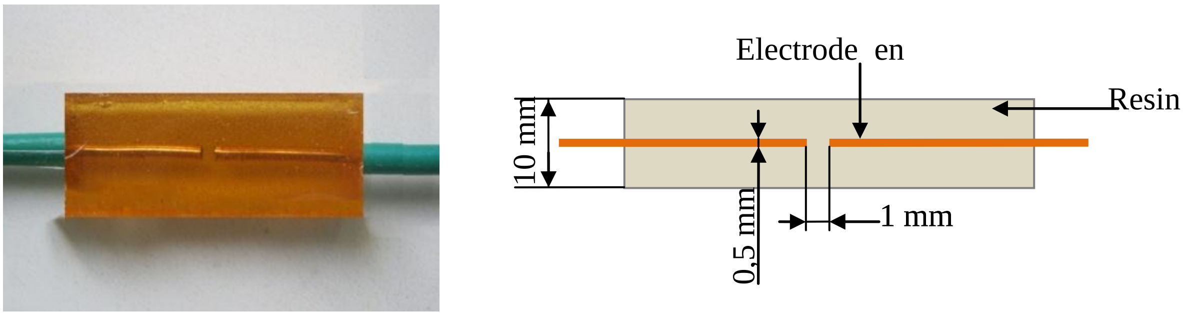

Figure 3 - The samples geometry for dielectric measurements

5. Results and discussion:

Dielectric spectroscopy analyzes allow to observe the evolution of the dielectric properties of the unfilled resin and loaded on the frequencies of 1 KHz and 10 KHz depending on the conditioning temperature of 5 ° C to 150 ° C and a humidity 60% RH.

Figures 4 and 5 respectively represent the mean curves of changes in the electrical resistance of the samples loaded and unloaded. Curves of these two samples show that the measurements are reproducible during the temperature ramp across the frequency range but with a little more dispersion at low temperatures.

Figure 4 shows the changes in the absolute values of the resistances of four types of samples during packaging at a frequency of 1kHz. The sharp decrease in resistance of the samples comb can be attributed to a low resistance between the electrodes at the interface charge / resin. This is due to a highly conductive resin phase-inter degraded by temperature.

The resistance of the reference samples (matrix) before packaging is identical in the presence of charges and k is approximately 150,106 Ω. From Figure 4, the resistance decreases during packaging. However, the drop is larger for samples not loaded.

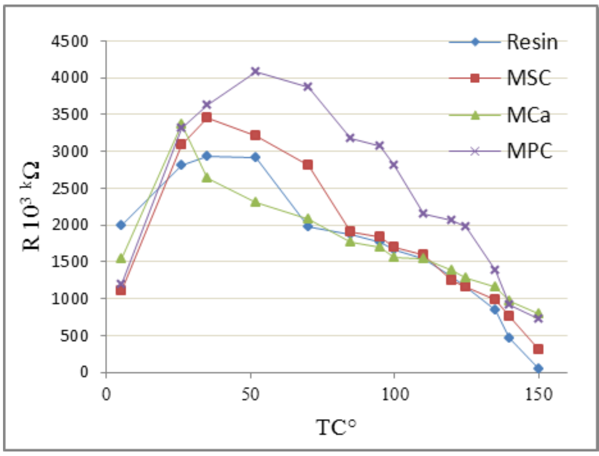

Figure 4 - Variation of the electric resistance of the various formulations depending on temperature at a frequency of 1kHz conditioning

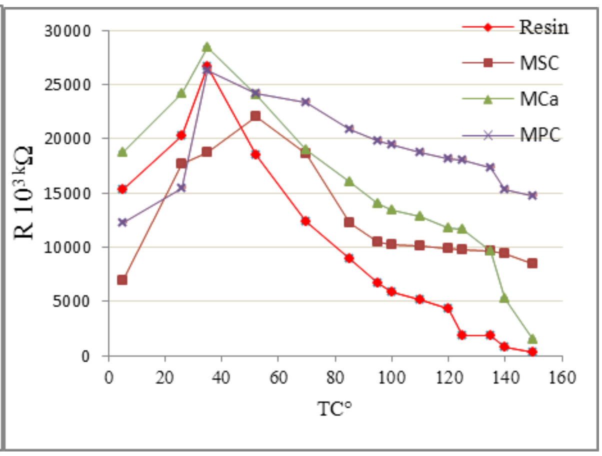

Figure 5 - Variation of the electrical resistance depending on temperature conditioning at a frequency 10 kHz

After a few days of conditioning, the resistance reaches the maximum at a temperature of 50 ° C, and that for all compositions where there was 26·106 kΩ for the composition (MPC), 28,4·106kΩ for the composition (MCa) 22,1·106 kΩ for (MSC) and finally 28,5·10^6kΩ for the matrix, the latter therefore a very low resistance and very rapid degradation vis-à-vis the conditioning temperature, unlike (MCa) shows that even very remarkable resistance at elevated temperatures.

After about 70 ° C, the resistance of the samples fall. Curves (MSC), (MCa), (MPC) in Figure V shows that the drop in resistance is much higher for the unfilled matrix, unlike others charged remain in their minimum value of 1·106 Ω.

The curves shown in Figure V represent the electrical resistance of the same samples with a frequency of 10kHz.

Curves representative's variation of the electrical resistance show that the measurements are reproducible during the temperature ramp over the entire frequency but with a little more drop at high temperatures. Behavior is proportional to the first category of tests at a frequency of 1 kHz, which explains the influence of the frequency is quite low on behavior, and that the load and its nature, and temperature are the bills major changes, and this time it is the composition that shows a threshold MPC remarkable contribution to other compositions and remains the same as that tested at a frequency of 1 kHz, where we recorded 26·106 kΩ at a temperature of 70 ° C. Unlike the unfilled resin, which remains very low values, which clarified the influence of the load and its importance for the isolation of this type of composite.

The influence of the expense is recognized when compared with the unfilled resin and the temperature conditioning of the two values of frequencies tested. The results show that the resistance of the resin with different loads evolves similarly to maximum values comparable to results found in the literature.

The objective of these measures is to obtain reference values for the resin with coated electrodes, in order to justify measures resistance. The maximum values of resistance of a coating unloaded equipped with brass inserts to explain very clearly the influence of the frequency and type of load during packaging of these dielectric properties. The fall of these properties during packaging elucidates the degradation of coatings depending on the temperature, and to justify this, a microscopic study is performed on the samples before and after conditioning packaged measure.

6. Conclusion

Under constraints of temperature and humidity, there is a drop in dielectric properties under AC voltage. The optical microscope images show that this is due to a physical effect of degradation of unfilled resin and moisture build-up between the cracks at the interface of the electrode / matrix.

These observations contribute to the formulation of materials and design of hardware and highlight a mode dielectric breakdown may involve a number of insulation composites.
The fillers addition such as pozzolana and cullet, the epoxy resin gives a great interest in its use for the isolation of metal at high voltages, such as transformer windings used in electrical distribution or traction. The resin may be subjected to the influence of the external environment.

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