Effect of Nano Fillers on Electrical Performance of Epoxy Composite Insulators

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***Abstract*: This work illustrates a study the performance improvement of epoxy composites as insulation material at different concentration of filler loading. Epoxy composite disc samples have been prepared after incorporated with different concentration of inorganic fillers e.g. Titanium dioxide (TiO_2), silica (SiO_2) and different size (micro and nano) to improve the electrical properties in addition to maximize dielectric strength and increase the electrical strength of material. hybrid epoxy composite constructs from epoxy and two different filler in concentration and in size. The dielectric strength of epoxy composites and hybrid composites is larger than the dielectric strength of epoxy due to using the inorganic filler. Dielectric strength of micro composite, nano composite is 95.4 kV/mm and 100.8 kV/mm respectively using 1% micro silica filler or 0.5% nano silica filler. Nano hybrid epoxy composite loaded with two different types of filler has 97 kV/mm dialectical strength.**

***Index:* Epoxy composites, Electrical properties, Matrix composite, Dielectric strength.**

I. Introduction

All polymer properties are changed by incorporation of reinforcement or filler particles into hybrid composite. Main types of properties influenced by fillers and reinforcements are classified to the mechanical, electrical, thermal and optical properties. The mechanical behavior is measured on the strength, stiffness, hardness, wear resistance and others. The electrical properties are detected by surface and cross resistivity, loss factor, permeability and so on.

Some methods of polymer graded material preparation are classified by gravitational and centrifugal casing, spraying, pressing, selective laser sintering. The different amount and sort of fillers were designed and then fabricated by pressing, gravity and centrifugal casting methods. Properties such as electrical surface resistivity, magnetic induction, coefficient of friction were tested and analyzed. Additionally distribution of filler particles in polymeric matrix was investigated [1].

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Some of researchers concern with the calculation of relative Permittivity, propagation delay, dissipation and loss. The dielectric constant, loss, partial discharge and break down voltages are measured by using ultra wideband [2].

Polymer–matrix nano composites (PMNs) is considered an attractive family of composite materials in which the nano-meter-size reinforcing fillers are uniformly dispersed in the polymer matrix on a nanometer scale as co mpared to con-ventional phase-separated macrocomposites [3].

The effect of the insulasion of electrically conductive and non-conductive nanofillers in a state of the art epoxy gelcoat was studied [4].

The non-ceramic insulators have been researched and improved with adding some type of filler. The ceramic and composite insulators have been compared with respect to their various characteristics. The composite insulators are being accepted increasingly for use in outdoor installations by the traditionally cautious electric power utilities worldwide. The tremendous growth in the applications of non-ceramic insulators is due to their advantages over the traditional ceramic and glass insulators. These include light weight, higher mechanical strength to weight ratio, higher resistance, better performance in the presence of heavy pollution in wet conditions, and better withstand voltage than porcelain or glass insulators [5].

The epoxy is insulating material but the filler must be added to epoxy for improving the physical and mechanical properties of insulator. There are different fillers can upgrade the mechanical properties [6].

The mixture between epoxy and microfiller is prepared in lab using mixer at high speed to get microcomposite while nanocomposite is manufactured by mixing at high speed and facing to magnetic or ultrasonic at the same time of mixing [7].

The aim of this work is to make an improvement to the electrical performance of epoxy composite insulators using different size fillers. Two types of inorganic fillers have been used such as; titanium dioxide (TiO_2) and Silica (SiO_2). Particle size of each filler in order of micro and nano scales. Also, it is focused on trying to find an appropriate percentage and size of inorganic filler adding to epoxy insulators which produces enhancements within dielectric strength (kV/mm).

II. EXPERMINTAL SET-UP AND TECHNIQUES:

A. Material Specimen

- *Materials*

Kemapoxy 150 JM is a two component solvent free liquid resin purchased from “Modern Chemicals for Modern

Building," Giza, Egypt. Component A is a medium viscosity liquid (epoxy resin) and Component B (hardener) is a difunctional primary amine.

The fillers used were titanium dioxide (TiO2) and silica (SiO2) in micro and nano size and supplied from Sigma-Aldrich, Steinheim, Germany. The micro size range was less than 20 µm and the nano size particles was less than 100 nm.

- *Curing of Epoxy Resin*

A mixture of the two components of Kemapoxy 150 JM, A and B in the ratio of (3:1 wt/wt %) were mixed thoroughly using a magnetic stirrer for 3 minutes. The mixture was degassed under vacuum to remove air bubbles, and then poured into a plastic mold. Curing occurred after 24 hours at room temperature (25˚C ±1). The sample is stable to machine after 7 days.

- *Preparation of Epoxy Composites*

Epoxy composites were prepared by mixing different ratios of each filler, titanium dioxide (TiO2) and silica (SiO2), in micro and nano size (0.1 up to 7% by weight) with epoxy/hardener mixture (3:1 wt/wt).

Also Epoxy composites were prepared from hybrid fillers (different types and concentration of titanium dioxide (TiO2) and silica (SiO2).The prepared samples of appropriate disc shapes (50 mm diameter and 0.5mm thickness) were molded and left to cure at room temperature overnight.

The coding of epoxy composite samples at different type and concentration of fillers is given in table (I).

Table (I)

Code of epoxy composite samples at different type and concentration of fillers.

a) *For microcomposites and nanocomposites.*

Concentration of filler (Wt. %)	Filler Type			
	Micro TiO_2	Nano TiO_2	Micro SiO_2	Nano SiO_2
0.1	$Ti_{M0.1}$	$Ti_{N0.1}$	$Si_{M0.1}$	$Si_{N0.1}$
0.25	$Ti_{M0.25}$	$Ti_{N0.25}$	$Si_{M0.25}$	$Si_{N0.25}$
0.5	$Ti_{M0.5}$	$Ti_{N0.5}$	$Si_{M0.5}$	$Si_{N0.5}$
0.75	$Ti_{M0.75}$	$Ti_{N0.75}$	$Si_{M0.75}$	$Si_{N0.75}$
1	Ti_{M1}	Ti_{N1}	Si_{M1}	Si_{N1}
3	Ti_{M3}	Ti_{N3}	Si_{M3}	Si_{N3}
5	Ti_{M5}	Ti_{N5}	Si_{M5}	Si_{N5}
7	Ti_{M7}	Ti_{N7}	Si_{M7}	Si_{N7}

b) *Hybrid composites*

Filler size and concentration %	Filler type		
Nano 0.05/Nano 0.05	TiO_2/ SiO_2	TiO_2	SiO_2
Nano 0.375/Nano 0.375	$Ti_N/Si_{N\ (0.1)}$		
Micro 0.05/Nano 0.05	$Ti_N/Si_{N\ (0.75)}$		
Micro 0.375/Nano 0.375	$Ti_M/Si_{N\ (0.1)}$	Ti $_{M/N\ (0.1)}$	Si $_{M/N\ (0.1)}$
Nano 0.05/Micro 0.05	$Ti_M/Si_{N\ (0.75)}$	Ti $_{M/N\ (0.75)}$	Si $_{M/N\ (0.75)}$
Nano 0.375/Micro 0.375	$Ti_N/Si_{M\ (0.1)}$		
Micro 0.05/Micro 0.05	$Ti_N/Si_{M\ (0.75)}$		
Micro 0.375/Micro 0.375	$Ti_M/Si_{M\ (0.1)}$		

Testing samples were performed by using hand tools to format the sample as in figure (1).

The dimensions of the samples were limited due to the limitations of the capabilities of ac supplies available in the high voltage laboratory.

a b

Figure (1): Disc sample.

A. The sample after complete curing.
B. The sample with 0.5 mm thickness and 50 mm diameter.

B. *Test Apparatus*

Puncture Test Supply and Electrodes Final is illustrated as following. The ac high voltage was obtained from a single phase high voltage transformer (100 kV-5KVA). The output voltage of the transformer is a smoothly controlled by (0-220V) variac regulating the voltage applied to its primary winding. Two similar copper sphere electrodes of diameter 36mm and tip radius is 25mm are designed in mushroom profile as shown in figure (2). The electrodes are fixed in clear polyester tank of oil with position in the center of disc sample and also have diameter greater than thickness sample with 10 times [9].

Figure (2): Composite sample which is fitted between mushroom electrode and immersed in oil.

Puncture test is most commonly used to determine the required breakdown voltage to pass the current through the thickness of a test specimen (puncture). This test is also used to determine dielectric strength of material. It may also be used to determine dielectric breakdown voltage along the interface between a solid specimen and a gaseous or liquid surrounding medium to prevent the discharges and flashovers on the surface of sample according to ASTM D 149.

The voltage was gradually increased at an almost constant rate of 0.5 KV/sec until the breakdown occcurs using test arrangement as in Figure (3). The breakdown voltage is defined as the maximum voltage that the insulator is punctured and large arc current is built between the electrodes through the sample.

The specimen is fitted across the high voltage at high voltage lab as in figure (4).

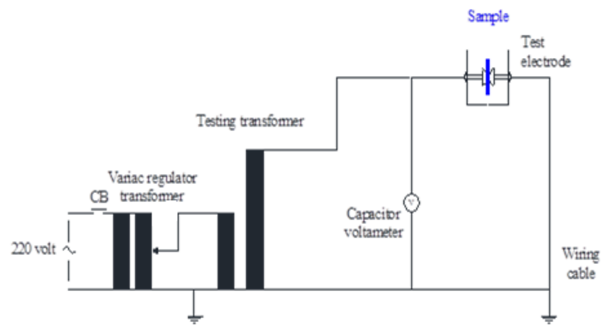

Figure (3): Schematic diagram of puncture test arrangement.

Figure (4): High voltage arrangement for determine break down test

C. *Measurement and uncertainty*

The result of a measurement is only an approximation or estimate of the true value of the specific quantity subject to result measurement.

The sources of uncertainty in the results of a measurement can be affected by many factors and some considerations as following:

- Reference standards and measurement equipment
- Measurement Setup
- Measurement Process
- Environmental Conditions

Misreading of instrument results, incorrect adjustments, using the wrong instrument, errors in recording calibration data, and computational errors is found practical. All of these errors can be avoided with proper training and attention to detail [10].

All identified standard uncertainty components, whether evaluated by Type A and Type B methods, are combined to produce an overall value of uncertainty to be associated with the result of the measurement, known as the combined standard uncertainty[11].

In this case the input estimate x_i is usually the arithmetic mean or the average of the individual observed values that is given by:

$$x_i = \bar{X} = \frac{1}{n}\sum_{k=}^{n} x_{i,k} \tag{1}$$

$$u(x_i) = s(\bar{X}_i) = \frac{s(X_i)}{\sqrt{n}} \tag{2}$$

Where:

$$s(X_i) = \sqrt{\left(\frac{1}{(n-1)}\sum_{k=1}^{n}\left(x_{i,k} - \bar{X}_i\right)^2\right)} \tag{3}$$

When the number of repeated measurements is low (n<10), the type (A) evaluation of standard uncertainty, as expressed by equation (4.2), should be multiplied by the student,s factor (t) obtained from table II in terms of number of samples n for a required confidence level p.

$$U(x_i) = t \times s(\bar{X}_i) = t \times \frac{s(X_i)}{\sqrt{n}} \tag{4}$$

If the probability distribution characterized by the measurement result y and its combined standard uncertainty u(y) is approximately normal (Gaussian), and u(y) is a reliable estimate of the standard deviation of y, then the interval [y-u(y)] to [y+u(y)] is expected to encompass approximately 95% of the distribution of values that could reasonably be attributed to the value of the quantity Y of which y is an estimate. This implies that it is believed with an approximate level of confidence of 95% that Y is greater than or equal to [y-u(y)], and is less than or equal to [y+u(y)], which is commonly written as Y=y±u(y) [10].

Although the combined standard uncertainty u(y) is used to express the uncertainty of many measurement results, for some commercial, Industrial, and regulatory applications (e.g., when helth and safety are concerned), what is often required is a measure of uncertainty that defines an interval about the measurement result y within which the value of the measured Y can be confidently asserted to lie. The measure of uncertainty intended to meet this requirement is termed expanded un certainty, suggested symbol U, is obtained by multiplying u(y) by a coverage factor, suggested symbol k.

$$U = k \times u(y) \tag{5}$$

It is confidently believed that Y is greater than or equal to [y-U], And is less than or equal to [y+U], which is commonly written as [Y=y±U]. In general, the value of the coverage factor k is chosen on the basis of the desired level of confidence to be associated with the interval defined by the equation (4.5).

III. RESULTS AND DISCUSSION:

A. Electrical insulation properties for epoxy composites at different type and size of fillers.

Electrical insulation properties of epoxy composites are explained from studying the breakdown voltage test. The breakdown test is named with puncture test referring to the damage of sample after the electrical test by passing the current through the sample which becomes punctured.

The breakdown test is considered destructive test because the specimen is punctured by passing current through sample between two electrodes. The physical view of specimen after breakdown is repeated for all specimens as shown in figure (5). The dark area is polluted due to carbon particle which is found by decomposition the oil around the specimen during test as in some specimens at figure (5).

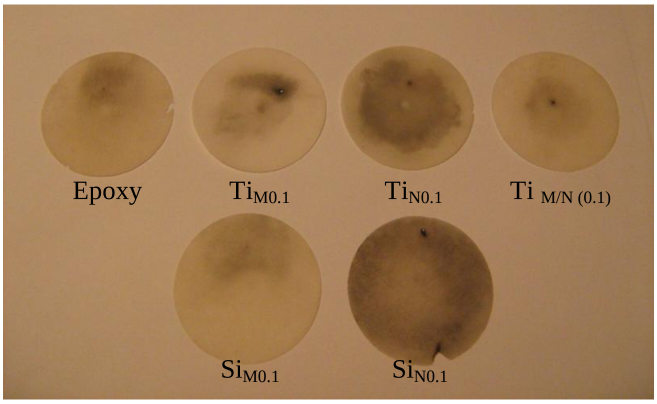

Figure (5): Breakdown voltage of epoxy and composites at different filler type

B. Benefit of uncertainty concept in the breakdown voltage test

The breakdown test is applied for polymer composite to explore the dielectric strength of composite material. The electrical test is repeated to four samples with the same chemical construction. The uncertainty concept is used to detect the error in reading and how this result to be applicable and error within range. The error U is calculated with the last equations (1), (2), (3), (4) and (5). If the number of similar samples n will be equal to four and also student factor $\rightarrow$k , the coverage factor (k) will be 3.18 at confidence level 95%. The following table (II) illustrates the entire step in calculation of error to find U in value and percentage.

The error percentage not exceed 5% in the most readings of breakdown for samples which means that the confidence level become 95%.

There is four samples with the same chemical feature, the average of breakdown for this samples represent the mean advised value for this type and also has error U in form BDV=BDV±UBDV. For uncertainty principle, the assumption of confidence level in 95% is suitable with the final results in error at range 5%.

The uncertainty of the last data gives the convergence level between the four tested data which give your tension that the breakdown data for tested samples are correct and reasonable.

The following table shows that the error in result within 5% range when the test is applied for different specimens with same chemical properties.

Table (II) The calculation of uncertainty for microcomposite and nanocomposite titanium dioxide specimens

Samples	Avarge	$u(x_i)$	k	U	U%
Epoxy	42.08	0.65	3.18	2.06	4.89
$Ti_{M0.1}$	45.23	0.32	3.18	1.01	2.23
$Ti_{M0.25}$	46.95	0.44	3.18	1.40	2.98
$Ti_{M0.5}$	45.68	0.70	3.18	2.22	4.86
$Ti_{M0.75}$	46.53	0.53	3.18	1.68	3.61
Ti_{M1}	44.85	0.49	3.18	1.57	3.50
Ti_{M3}	42.50	0.42	3.18	1.32	3.12
Ti_{M5}	41.33	0.56	3.18	1.78	4.30
Ti_{M7}	42.70	0.36	3.18	1.15	2.70
$Ti_{N0.1}$	50.83	0.58	3.18	1.85	3.63
$Ti_{N0.25}$	46.55	0.69	3.18	2.20	4.72
$Ti_{N0.5}$	37.70	0.26	3.18	0.84	2.23
$Ti_{N0.75}$	37.18	0.44	3.18	1.41	3.80
Ti_{N1}	36.25	0.46	3.18	1.45	4.00
Ti_{N3}	35.93	0.64	3.18	2.02	5.63
Ti_{N5}	36.63	0.74	3.18	2.35	6.42
Ti_{N7}	33.83	0.46	3.18	1.47	4.35

C. Breakdown Test for Epoxy Composite using Silica filler

The silica is considered insulation material and is mixed with epoxy resin to format epoxy composites with different electrical properties. The silicone dioxide filler is added to epoxy with two different particle size in nano and micro meter.

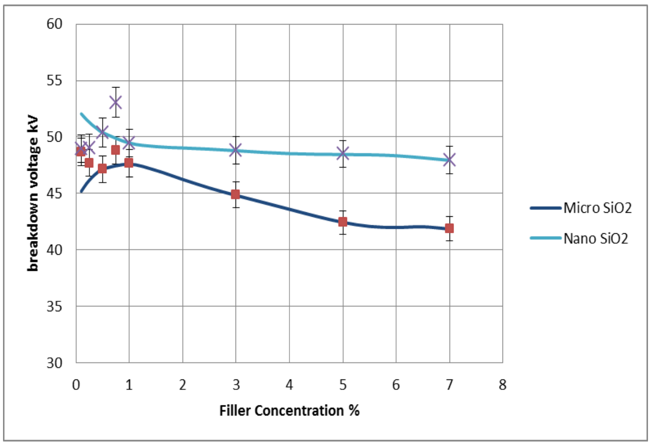

Figure (6): Breakdown voltage of epoxy composites at different concentration of micro and nano silica filler.

When the filler concentration increase for nano silica composite, Breakdown voltage will be decreased with small variation. When the filler concentration increase from 5% to 7% for micro silica composite, breakdown is fixed at 42 kV approximately as in figure (6).

The increase of filler leads to decreasing the breakdown voltage because the epoxy composite is saturated with filler and is not effective by raising the filler concentration.

D. Breakdown Test for Epoxy Composite using Silica filler

The epoxy has physical structure which contains some spaces. This space is filled with filler regardless of material type.

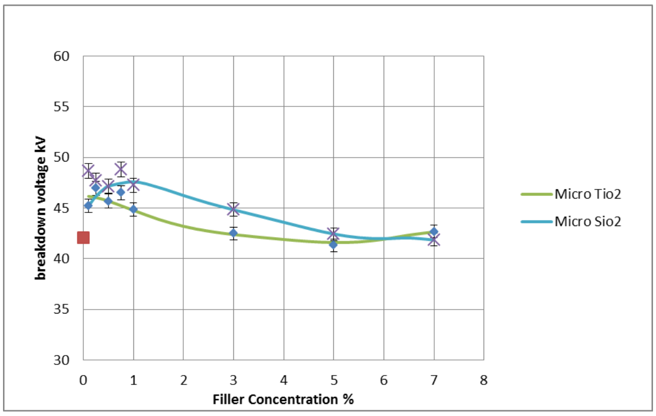

Figure (7): Effect of filler concentration of micro titanium dioxide and silica on the breakdown voltage for epoxy composites.

Same trend can be observed for breakdown voltage against the filler concentration between microcomposite loaded with titanium dioxide and microcomposite loaded with silica dioxide as in figure (7).

The difference between breakdown voltage values for micro filler between the titanium dioxide and silica dioxide is very small. The filler type is not important in the performance epoxy composites. The particle size of filler is responsible on modification the characteristic of epoxy composites.

The variance of breakdown for microcomposite under concentration above 1% and different filler type is very large comparing with microcomposite while filler concentration is smaller than 1% up to 0.25% as shown in figure (7).

E. Breakdown Voltage Test for Nanocomposite using different filler

The breakdown voltage of nano silica composite is larger than breakdown for nano titanium composite at any concentration as in figure (8).

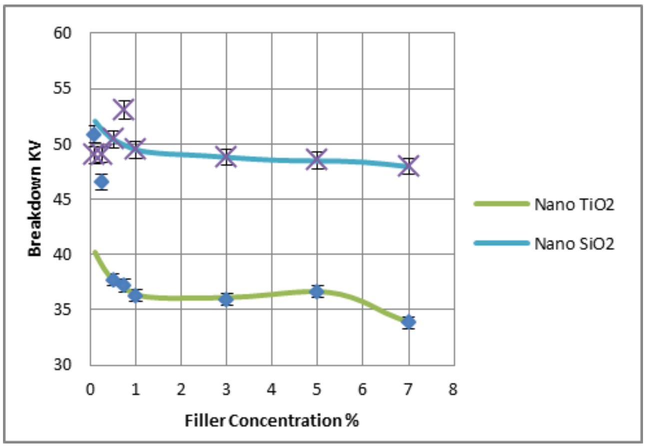

Figure (8): Effect of breakdown voltage for epoxy composites against filler concentration of micro titanium dioxide and silica.

The nano silica composite of 0.1% concentration records maximum breakdown voltage by comparing with other spicmen at different concentration as in figure (8). If the filler concentration of nano titanium composite varies from 1% to 5%, breakdown for composites is not change.

F. Electrical insulation properties for hybrid composites at different type and size of fillers

The electrical and mechanical performance for epoxy composites depends on the filler. Therefore the filler may be contains two types at different size of particle. The main topic of addition the filler improves the electrical performance and also for the physical and mechanical.

The following figures illustrate the effect of hybrid filler on the breakdown voltage for epoxy hybrid composites by changing the type, size and concentration of filler material.

The hybrid filler contains two different material in types or in particle size.

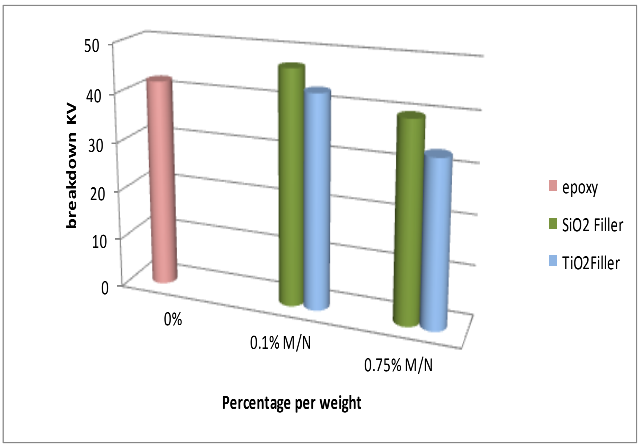

Figure (9): Variation in breakdown voltage of hybrid composites with micro and nano filler for different materials.

when the hybrid composite contains the filler material with micro and nano size, the breakdown voltage of composite will be decayed at increasing the filler concentration as shown in figure (9).

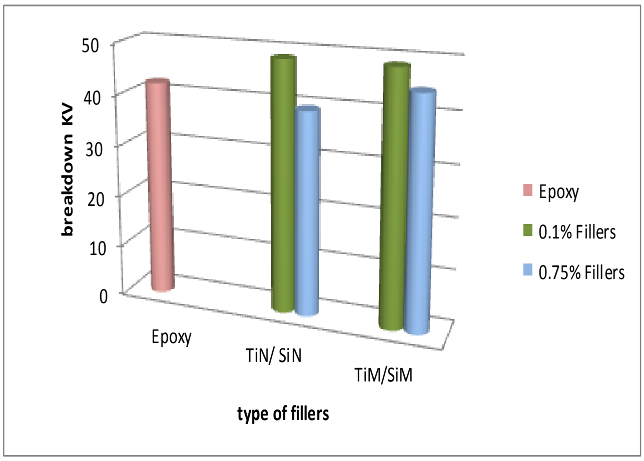

Figure (10): Variation in breakdown voltage of hybrid composites with two different materials and same particle size at varied concentration.

When the hybrid composite contains two different filler material with the same particle size, the breakdown will be decayed at 0.1% filler concentration by increase size of particle but the breakdown will be increased at 0.75% filler concentration at changing size of filler particle as shown in figure (10).

IV. Conclusion

Electrical properties for epoxy composite filled with Silica is reliable at 0.1% nano particle size and has dielectric strength better than micro silica composite at the same concentration. The silica nano filler which is used in composite give maximum dielectric strength 103 kV/mm where the epoxy give maximum dielectric strength 84 kV/mm. Dielectric strength of hybrid composites loaded with micro titanium dioxide and micro silica records 97.35 kV/mm when the dielectric strength of microcomposites has 95.2kV/mm, 91.8kV/mm for silica and titanium dioxide filler respectively. Hybrid composite filled with micro and nano silica filler has dielectric strength 94 kV/mm. Finally, the nano silica filler improve the electrical properties of epoxy composites and increase the dielectric strength of composites.

References

[1] J. Stabic, A. Dybowska and M. Chomiak ,"Polymer composites filled with powders as polymer grade materials", journal of Achievements in materials and manufacturing engineering. Vol. 43,2010 pp.153-161.

[2] M.G. Mayes, J.R. Mayes and M.B. Lara, "High Voltage Properties Of insulating Materials Measured in the Ultra Wideband", Pulsed, PP. 954 – 957, DIO: 10.1109/PPC.2005.300452.

[3] Xiaowu Fan, Lijun Lin and Phillip B. Messersmith,"Surface-initiated polymerization from TiO2 nanoparticle surfaces through a biomimetic initiator: A new route toward polymer–matrix nanocomposites", elsevier, composites science and technology vol. 66 (2006) pp 1195–1201.

[4] P. Karapappas, P. Tsotra and K. Scobbie, "Effect of nanofillers on the properties of a state of the art epoxy gelcoat", Express Polymer Letters Vol. 5, No.3 (2011) pp218–227.

[5] Gençoğlu, M.T. "The Comparison of Ceramic And Non ceramic Insulators", e-Journal of New World Sciences Academy Natural and Applied Sciences, 2, (4), A0038, 274-294.

[6] Y. X. Zhou, P. X. Wu, Z-Y. Cheng, J. Ingram and S. Jeelani "Improvement in electrical, thermal and mechanical properties of epoxy by filling carbon nanotube", DOI: 10.3144/express Polymer Letters Vol.2, No.1 (2008) PP. 40–48.

[7] Muneaki Kurimoto, Hitoshi Okubo, Katsumi Kato, Masahiro Hanai, Yoshikazu Hoshina, Masafumi Takei and Naoki Hayakawa,"Permittivity Characteristics of Epoxy/Alumina Nanocomposite with High Particle Dispersibility by Combining Ultrasonic Wave and Centrifugal Force", dielectrics and electrical insulation, IEEE transactions on Volume:17, Issue: 4), PP.1268 - 1275 , DIO: 10.1109/TDEI.2010.5539699.

[8] Titanium dioxide (TiO_2) nanoparticles, www.kolektor.si.

[9] Sung-Hoon Cho, Yu-Min Kim, Jung-Hun Kwon, Kee-Joe Lim, Eui-Hwan Jung, Hung-Kyu Lee and Pan-Seok Shin, "Improvement of Electrical and Thermal Characteristics of Nano-Micro Epoxy Composite", transactions on electrical and electronic materials , Vol. 12, No. 4, pp. 160-163, August 25, 2011.

[10] Edward Brown, Quality Systems Lab, Boca Raton, FL 33487" Understanding and Expressing Measurement Uncertainty associated with DC and Low Frequency Metrology" http://www.pjlabs.com.

[11] Keith Birch, "Estimating Uncertainties in Testing", N C Barford, "Experimental Measurements, Precision, Error and Truth", Addison-Wesley Publishing Company, Inc, London.

[12] EUROLAB Technical Report 1/2006, "Guide to the Evaluation of Measurement Uncertainty for Quantitative Test Results", www.eurolab.org.