Improvement Thermal and Electrical Properties for System Epoxy- Al_2TiZrO_7 Nanoparticles

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Abstract- In this research the ceramic system (Al_2TiZrO_7) was prepared using the effective mechanical mixing method by 1:1:1mol per gram .The materials were mixed with a granular size of about 35nm. The epoxy used in this work type 105 is a transparent liquid with a density of 1.19 g/ cm^3 and a high viscosity and good thermal and electrical insulation. The samples were prepared using the liquid and the ultrasound technique. The ceramic powder was mixed with the epoxy by weight (3% , 5% , 7% and 10%) . Samples are formed using silicon molds according to standard specifications. Thermal tests were performed using a device (Thermal Conductivity analyzer TCi), the rustles showed a significant improvement in thermal transfer of the composites compared to the net sample (epoxy). The electrical tests were performed using the LCR – Meter with a frequency range of (50Hz – 5MHz) , the results showed a significant reduction in the electrical resistivity values by increasing frequency , the electrical conductivity was calculated at 1 MHz The results interpreted based on the practical density and the electron microscopy, as well as the energy dispersion spectroscopy.
Keywords: ceramic system, effective mechanical mixing, ultrasound technique, thermal transfer, electrical conductivity.

1. Introduction

Epoxy-based polymers are widely used in applications ranging from microelectronics to the aerospace industry.Their insulating nature can cause accumulation of electro- static charge on their surface, causing local heating and premature degradation to the electronic components or space structures. They can be made electrically and thermally conducting by the addition of various types of reinforcements (nano or micro)[1,2]. Demand for advanced materials with better properties has been increasing to meet new requirements or replace existing materials. Polymer composites have superior strength, stiffness, toughness, hardness, and heat distortion temperature compared to metal material properties. Recently, polymer composites have been studied for a wide range of applications including bio-tech, automotive, and aerospace industries [3]. Because composites made of epoxy-based materials furbish outstanding corrosion resistance, electric insulation, thermal stability, dimensional stability, and durability, epoxy resins are widely used as polymer matrices for advanced composite materials [4-7]. Although epoxy resin has outstanding properties, it has weak strength and brittle properties relative to other resins. To address the problems mentioned above, using an additional filler to strengthen the properties of epoxy resins has become a common practice. More specifically, micro- and nano-scaled fillers have been considered as filler materials for epoxy to realize high-performance composites. The properties of filler materials such as carbon nanotubes (CNTs), fibers, minerals, ceramic, and clay are exploited to compensate the weak and brittle properties of epoxy resin and also to address the issue of its low availability [8-10]. The addition of $AlTiZrO_7$ into polymers at very low volume could lead to considerable increase in their mechanical properties [11], electrical properties [12], thermal conductivity [13] and flame disruptive [14]. The aim of research is to develop a system of polymer – ceramic with thermal and electrical conductivity applicable in the field of electronic engineering.

2. Theoretical Part

Heat transport at the nana scale is a very interesting and technologically important area. With the reduction of object size, phonon modes and phonon densities of states change drastically, resulting in unusual thermal transport phenomena in microscopic systems. The thermal conductivity of the Nan belts is significantly suppressed in comparison to that in the bulk due to increased phonon–boundary Scattering an modified phonon dispersion [15]. This size effect can lead to localized heating in Nan electronics [16], The thermal conductivity can be calculated using the equations, [17].

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$Q = \frac{dH}{dt} = -\frac{\lambda A dT}{dx}$.......(1)
Where:
Q: The Heat flow per time (Watt),
H = Heat (J)
t = Time (sec)
λ = Thermal conductivity (W/K. m)
T = Temperature (K)
x = Height of test specimen (m)
A= cross sectional area of test sample (m^2)
The effusivity equation by ($Ws^{½}$ / m^2 . K)
$\varepsilon. effusivity = \sqrt{\lambda . \rho . Cp}$...................(2)
The thermal diffusion equation (m^2/ s):
$\delta = \frac{\lambda}{Cp . \rho}$..................... (3)
Where:
C_p: Specific heat capacity by (J/g. K)
ρ= Density of sample (g / cm^3)
The thermal Resistance equation by (m^2.K/ W)
$R_{thermal} = \frac{dX}{\lambda}$ ....(4).
The electrical properties of the neat and nanophase polymer blend were measured by using an Agilent 4294-A Precision Impedance analyzer. The composites were cut into disc with dimension of (20mm×4mm) (diameter, and thickness). The measurement was carried at frequencies from (50 Hz to 5 MHz) at room temperature. In the measurement, the impedance of the sample at each frequency was measured and recorded. The resistivity of the composites was calculated using the measured impedance and the geometry of the sample [18]. The impedance is given by the equations:
$Z = \frac{R}{\sqrt{1+(2\pi\ )^2 R^2 C^2}}$........................ (5)
Where *R* is the electrical resistance of a uniform specimen of the material measured in ohms (Ω,C) is the capacitor's capacitance and (*f*) is frequency. The resistance and capacitance are given by Equations (2-16) and (2-17):
$R = \frac{\rho.\ A}{V}$.............................. (6)
The electrical conductivity its inverse resistivity
$\rho = \frac{1}{\sigma}$....................................(7)

Where :(ρ) is the resistivity and (ε) is the permittivity of the dielectric material. Both of them are material parameters, A, V are area, and volume of sample, respectively [19].

3. Experimental and Materials used

Materials used in this study are listed as below:
1- Titanium oxide (TiO_2) Nano particles, nano powder size <25 nm> purity: 99.8 , 25gm HWNANO (China).
2- Zirconia oxide (ZrO_2) Nano particles, nano powder size <40 nm> , purity:99 25 gm. HWNANO (China).
3- Alumina (Al_2O_3) Nano particles, nano powder size <25 nm>, purity: 99 25 gm. HWNANO (China).
4-.Acetone Analytical reagent 0882- Thoas baker $(CH_3)_2CO$.
5- Epoxy resin 105 type. (Resin and Hardener) by weight (3:2).

In this study, the Preparation of ZrAl2Tio7 Molecular ratios (1:1:1) were selected in the preparation of this ceramic or system and according to the granular size of each powder (TiO_2(25nm), ZrO_2(40nm) and Al_2O_3(40nm)). After mixing the powders with the same method of effective mechanical mixing for two hours.then the reaction of the powders at a temperature(1000 c) in the tubular furnace for two hours and then was examined x-ray and examined SEM, preparation and development of the nanocomposites using the technique of diffusion or dispersion using the ultrasonic device at frequency 40kHz. Mix powder PMMA with powders (ZrO_2,Al_2O_3,TiO_2 and $ZrAl_2Tio_7$), using the method of mechanical mixing effective(AM) and this method is the use of balls weight(20gm) for each quantity of powder weight(5gm) of any proportion (2gm balls:5gm powders), and for (3 hours) and quickly(250rpm). after was put Monomer (chloroform) to the mixture by any proportion(5gm:3.5gm) of powder to the liquid and a period of (30 sec). then They shall be poured into special molds for examination and in standard dimensions according to the required examination.

4. Rustles and discussion

Table (1) and Figures (1, 2,3,4,5 and 6) summarize the practical results of the electrical conductivity test, where the relationship between the percentages of the material of the reinforcement and the coefficients were determined without the base material because their values are far from the values measured as is Clear the table. Electrical tests were carried out using a frequency impedance device ranging from 50 Hz to 5 MHz. The results showed a significant improvement in the values of capacitance, constant insulation and the electrical loss factor of all the reinforced materials compared to the epoxy base material. The reason is that the ceramic system has two important properties: the first is characterized by volumetric density, high surface area and the other advantage is the abundance of free electrons on its surface, The basis of the electrical conductivity values from $0.08x10^{-13}$ for the base material to $0.17x10^{-6}$ for ceramic-reinforced samples . In addition to the inverse relationship between resistance and frequency according to relation (5) of the theoretical part, this agreement with **reference** [20].

Table (2) and Figures (7,8,9,10, 11 and 12) The summary of the practical results of the thermal transfer properties of the base material (epoxy) and the composites prepared and reinforced by the nanocomposite system, where the technique (Thermal conductivity analyzer TCi) is shown. The results showed a clear improvement in the values of conductivity, flow, thermal propagation by increasing the percentage of the reinforcing material while the values of the capacitance and thermal resistance decreased by increasing the percentages due to the fact that the material has a thermal conductivity coefficient, density and relatively high surface area as well as the homogeneous distribution of the prepared ceramic powder where the figure (12) shows the image of the scanning electron microscope (SEM) and the spectrum of dispersion energy (EDx) to know the compounds involved in the composition of the compound. The reason for the low capacity and thermal resistance is the result of the inverse relationship with the flow and thermal diffusion as shown in Table (2), this is agreement with **reference** [21].

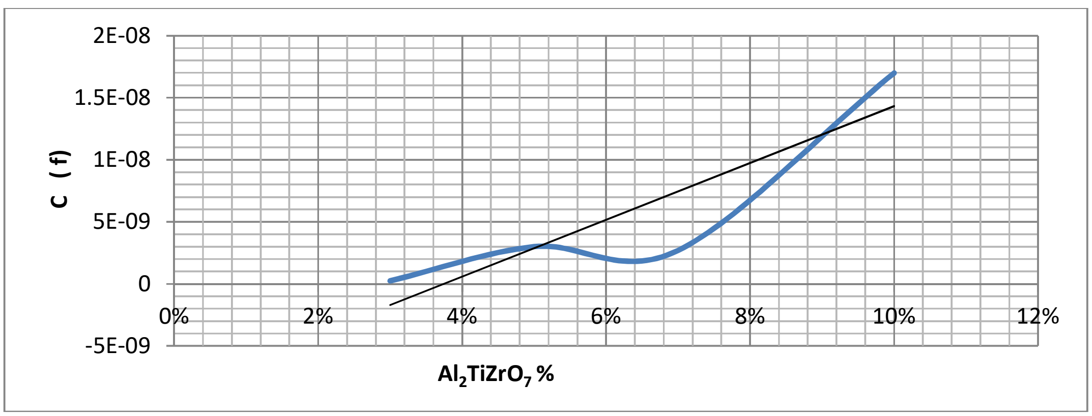

Fig.(1): Experimental of Electrical Capacitance at 1MHz.

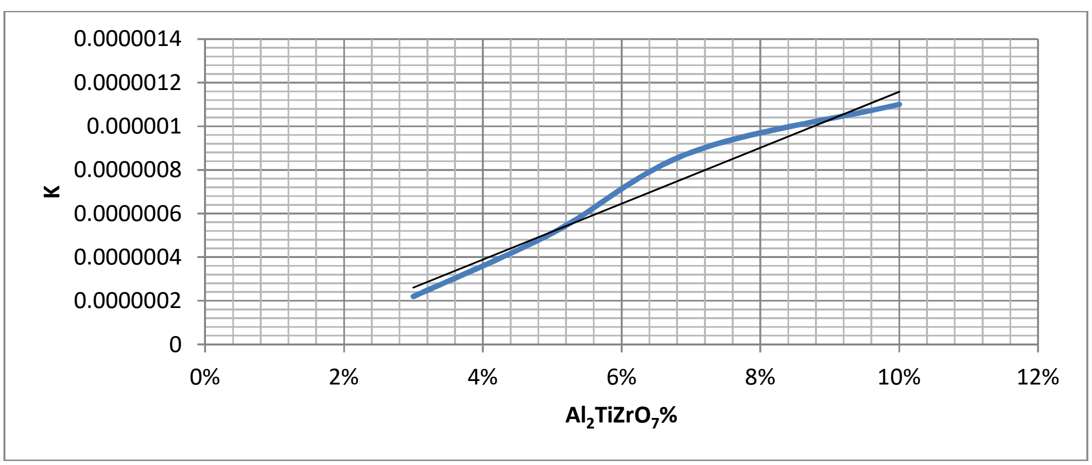

Fig.(2): Experimental of dialectical constant at 1MHz.

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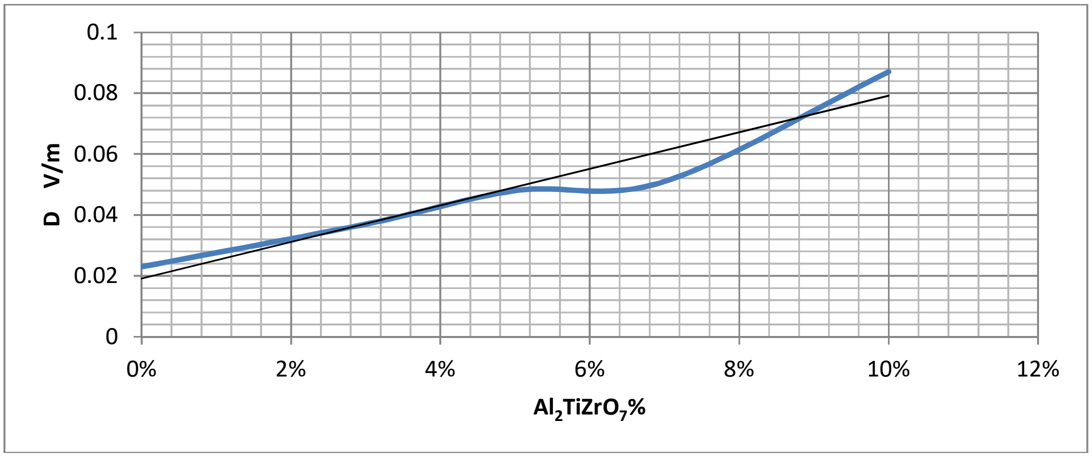

Fig.(3): Experimental of Loss Factor at 1MHz.

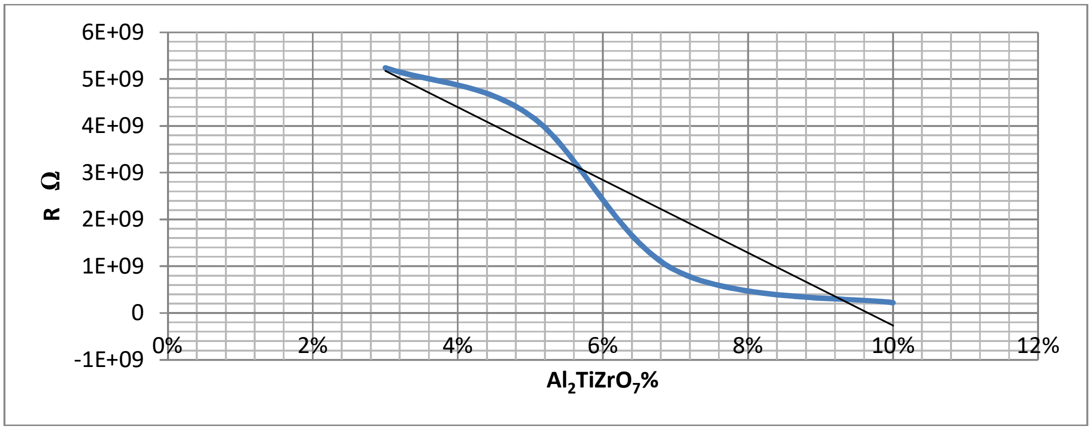

Fig.(4): Experimental of Electrical Resistance at 1MHz.

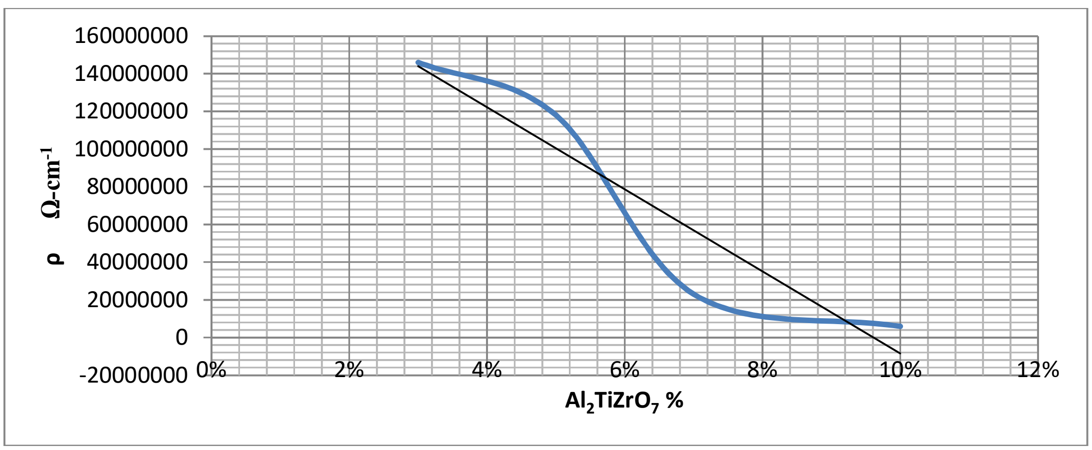

Fig.(5): Experimental of Electrical Resistivity at 1MHz.

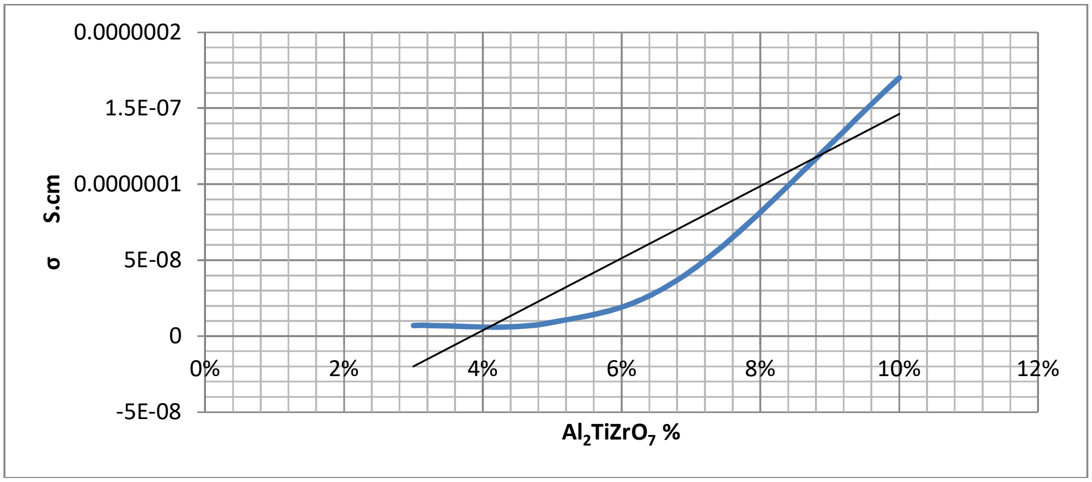

Fig.(6): Experimental of Electrical Conductivity at 1MHz.

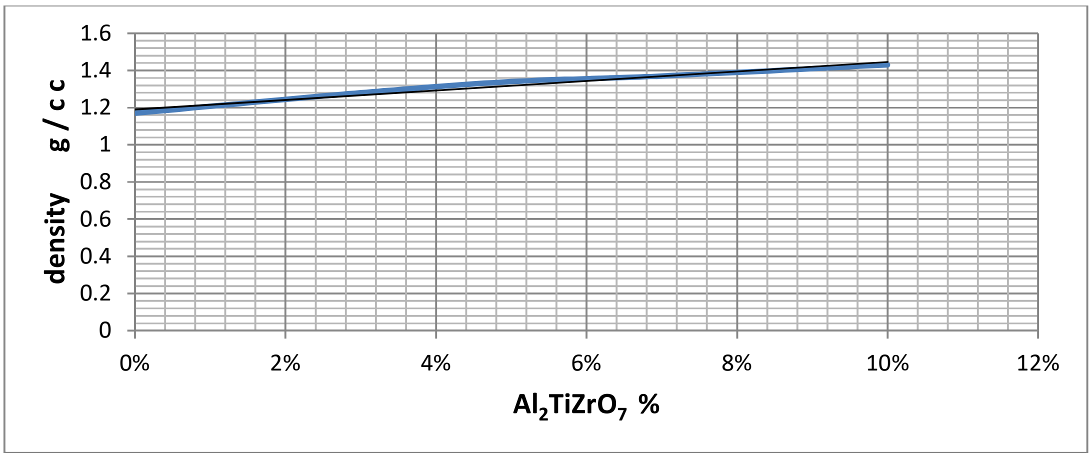

Fig.(7): Experimental density with percentage of Nano ceramic

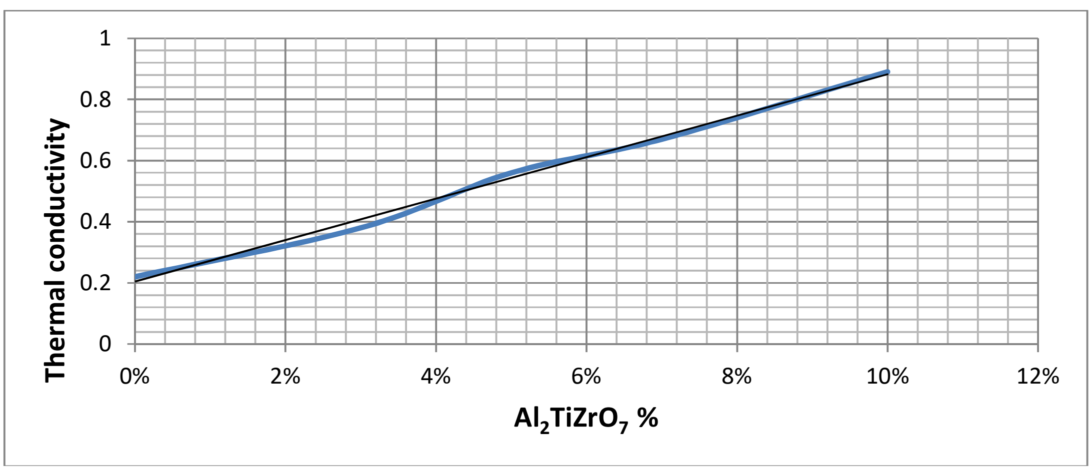

Fig.(8): Experimental Thermal conductivity with percentage of Nano ceramic

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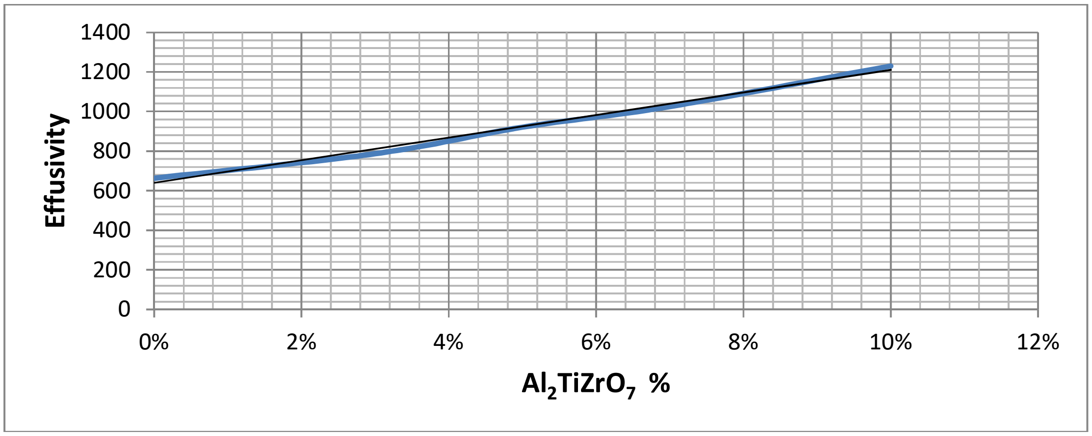


Fig.(9): Experimental Thermal Effusivity with percentage of Nano ceramic

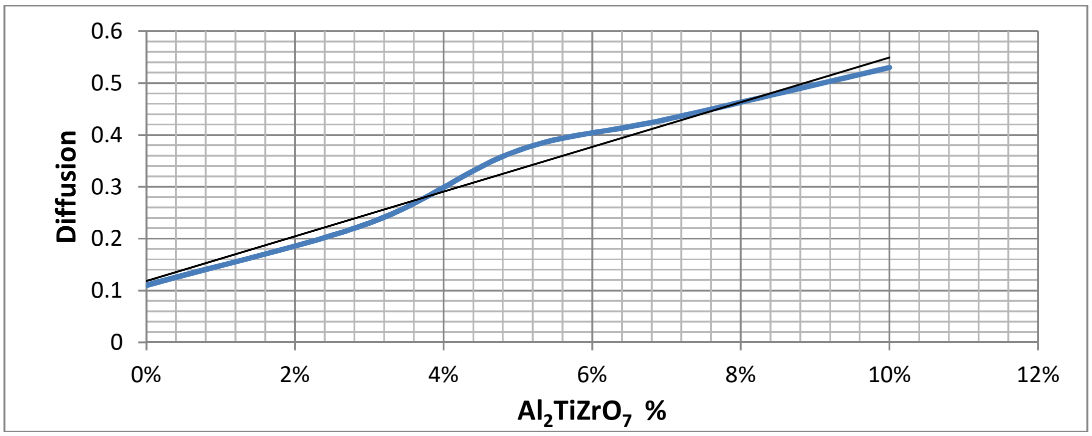


Fig.(10): Experimental Thermal Diffusion with percentage of Nano ceramic

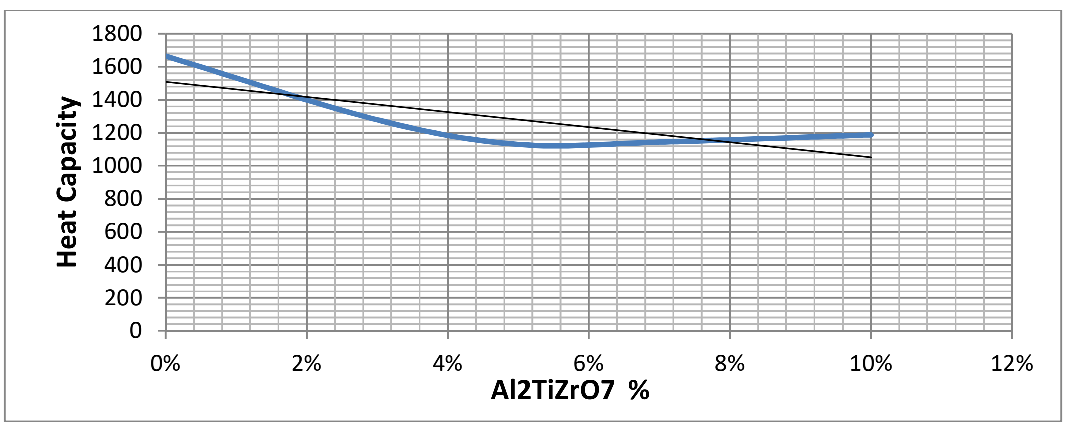


Fig.(11): Experimental Heat Capacity with percentage of Nano ceramic

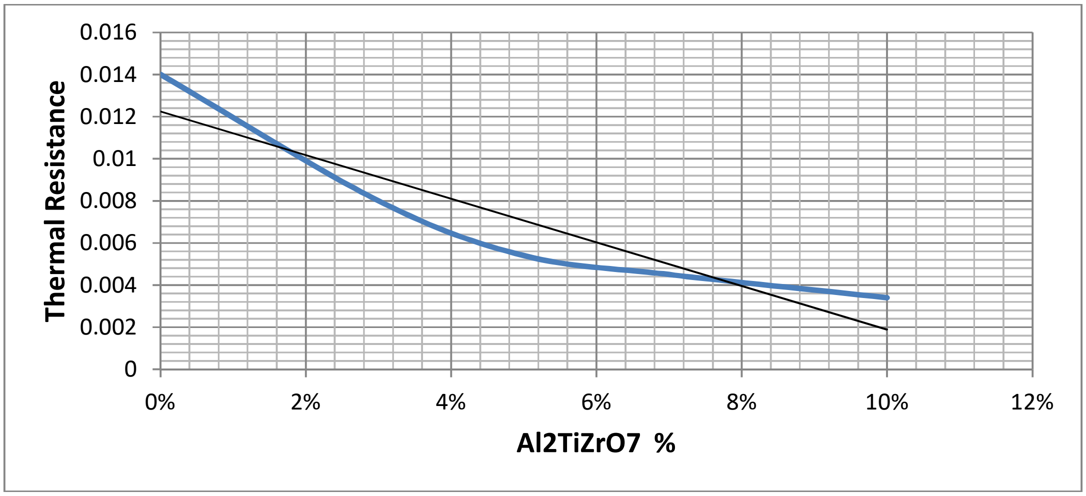
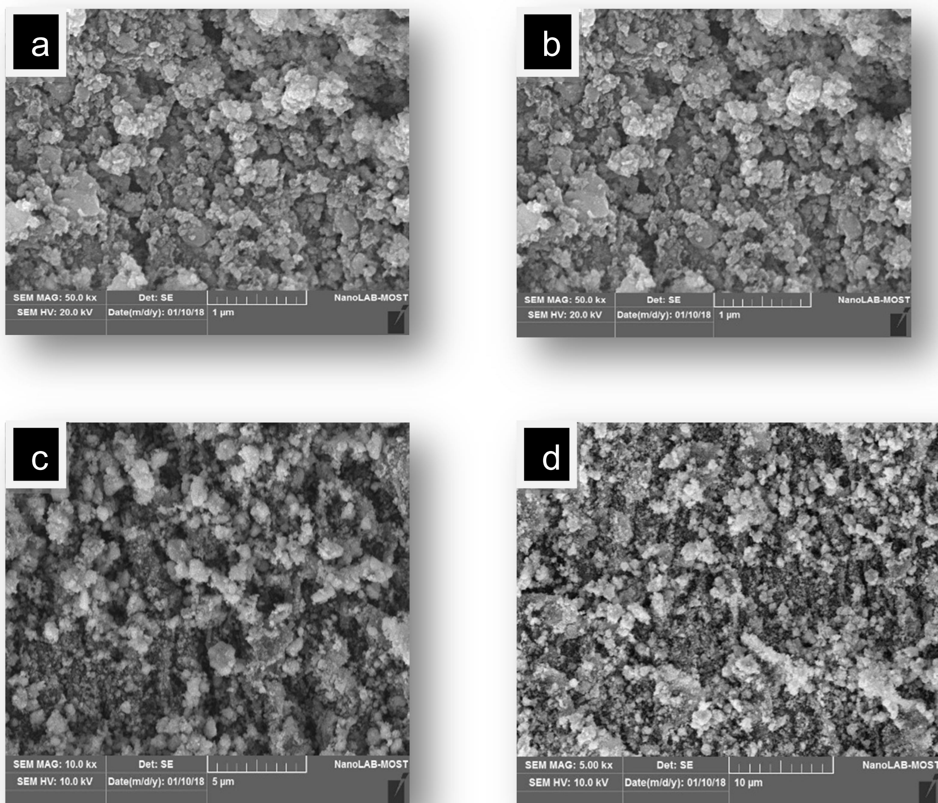


Fig.(12): Experimental Thermal Resistance with percentage of Nano ceramic.



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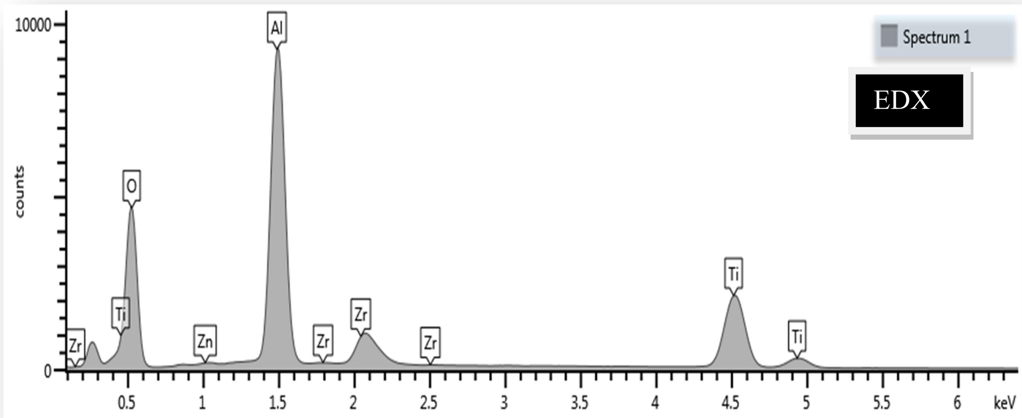

Fig.(13): (a,b,c,d) SEM micrographs and EDX

Table (1) Experimental of Electrical Properties at 1MHz

Sample	C (f)	K	D (V / m)	R (Ω)	ρ .cm)(Ω	б (S.cm^{-1})
0% $AlTiZrO_7$	11.0 x10^{-12}	0.4x10^{-8}	0.023	3910x10^{12}	12x10^{13}	0.08x10^{-13}
3% $AlTiZrO_7$	25 x 10^{-9}	0.22x10^{-6}	0.037	5240x10^{6}	146x10^{6}	0.007x10^{-6}
5% $AlTiZrO_7$	3.0 x10^{-8}	0.51x10^{-6}	0.048	4218x10^{6}	118x10^{6}	0.0085x10^{-6}
7% $AlTiZrO_7$	27x10^{-8}	0.88x10^{-6}	0.051	912x10^{6}	23x10^{6}	0.043x10^{-6}
10% $AlTiZrO_7$	17x10^{-7}	1.1x10^{-6}	0.087	220x10^{6}	5.9x10^{6}	0.17x10^{-6}

Table (2) Experimental Thermal Properties by TCi Analyzer

Sample	Density (g/ cm^3)	λ (w / m. K)	Ɛ ws^{½}/m^2.K	δ (mm^2 / s)	C_p (J/kg.K)	R cm^2.K.W^{-1})(
0% $AlTiZrO_7$	1.17	0.22	663	0.11	1665	0.014
3% $AlTiZrO_7$	1.28	0.38	787	0.23	1279	0.008
5% $AlTiZrO_7$	1.34	0.56	921	0.37	1130	0.0054
7% $AlTiZrO_7$	1.37	0.67	1025	0.43	1144	0.0045
10% $AlTiZrO_7$	1.43	0.89	1230	0.53	1188	0.0034

Conclusions

The most important conclusions reached:

1- Practical results of this research are the addition of the three system ceramic powder to the epoxy material, which is characterized by good thermal and electrical insulation, which led to its transformation intro a semi – conductive material, to some extent.
2-The rapid decrease in the capacitance values and thermal resistance of the composites and the added percentages compared to the net epoxy make these composites useful in the field of electronic applications in aircraft and spacecraft systems for their light weight and resistance to different conditions.
3- An important conclusion is that these composites can be considered a new class
Tri- system Nanocomposite

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