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Predictable Models and Experimental Measurements for Electric Properties of Polypropylene Nanocomposite Films

Ahmed Thabet*, Youssef Mobarak*, **
* Nanotechnology Research Centre, Electrical Engineering Department Faculty of Energy Engineering, Sahari 81528, Aswan University, Sahari City, 81528, Aswan, Egypt,
** Department of Electrical Engineering, Faculty of Engineering, Rabigh, King Abdulaziz University, Saudi Arabia

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ABSTRACT

This paper processed and characterized cost-fewer polypropylene (PP) nanocomposite films; an experimental work has been investigated for studying the electric properties of the new nanocomposite materials and compared with unfilled industrial materials in a frequency range up to 1 kHz. A small addition of nanoparticles (clay, and fumed silica) to polypropylene showed appreciable improvement in the electric reactance and conductance at different frequency up to 1kHz, in addition, an electric spectroscopy has been measured the electric properties of polypropylene with and without nanoparticles under variant temperatures (20°C, and 60°C). Cambridge Engineering Selector (CES) program were carried out the electrical/mechanical predictable models for the suggested materials. Finally, this paper leads to synthesize electrical insulating polypropylene nanocomposite films where the electrical properties are properly maintained in order to achieve more cost-effective, energy-effective and hence environmentally better materials for the electrical insulation technology.

Corresponding Author:

Youssef Mobarak,
Electrical Engineering Department Faculty of Engineering,
Rabigh, King Abdulaziz University, Saudi Arabia
Email: ysoliman@kau.edu.sa

1. INTRODUCTION

Polymer nanocomposite films have attracted wide interest with regard to enhancing polymer properties and extending their utility in recent years. The nanocomposite material which the nanoparticles are evenly distributed in the polymer material attracts attention as an insulating material because the properties of the original material can be drastically improved by adding a few percent of nanoparticles. Nano-composites represent a very attractive route to upgrade and diversify properties of the polymers. Nano-filler-filled polymers might be differentiated from micro-filler-filled polymers in three major aspects that the nano-composites normally contain smaller amounts, are in range of nanometers in size and have tremendously large specific surface area. All these characteristics are reflected in their material properties [1-4]. In general, fillers are added to polymeric materials in order to enhance thermal and mechanical properties. Over the past few years there have been few numbers of researches on the effect of fillers on dielectric properties of polymers [5-8].

The shift from ceramic electric insulating materials (e.g. porcelain and glass) and from oil-paper insulations to polymeric materials has been the major change in the field of high voltage insulation technology during the past three decades. Today polymers are widely used in most of the high voltage equipment, e.g. power transformers, insulators, capacitors, reactors, surge arresters, current and voltage sensors, bushings, power cables and terminations. The wide possibilities of the existing polymers and, particularly, the huge scenarios of new polymer composites in high voltage technology inspire the researchers of the field for innovative new materials and to study their properties [9-11]. Research work on novel polymer materials is a great significance both nationally and internationally in the field of power engineering, high voltage technology and environmental technology due to the increasing demands of more cost-effective, efficient, reliable and environmentally satisfactory high voltage equipment. Particularly, there is a need for developing a range of compact devices and accessories, for both outdoor and indoor conditions, in which novel and more reliable insulation systems will play the key role.

Nano-materials, in form of polymeric nano-composites, are foreseen as excellent candidates able to fulfill the new requirements. Nano-filler-filled polymers might be differentiated from micro-filler-filled polymers in three major aspects that the nano-composites normally contain smaller amounts, are in range of nano-meters in size and have tremendously large specific surface area. All these characteristics are reflected in their material properties [12-16]. Polypropylene PP is widely used as an insulating material for power cables. Electrical insulating polymers are usually modified with inorganic fillers to improve electrical, mechanical, thermal properties. Polypropylene is widely used as a dielectric insulation of power cable. Nanoparticles/polymer composites are now of considerable interest for their specific electrical properties. It is recognized that the interfaces between the host dielectric and the nanometric particles can strongly influence the dielectric properties of the composite material as a whole. Since interfaces dominate dielectric situations at this level, nanodielectrics and interfaces become inextricable. Low frequency polarization is a type of polarization concerning to interface polarization, and it strongly relates to the space charge storage and transportation in dielectric materials [17-21]. As of now, work is underway to examine the physical properties of nanocomposite materials composed of nanoparticles of metals and their compounds stabilized within a polymeric dielectric matrix [22-25]. The electric and optic properties of these materials have been demonstrated to be highly dependent on the size, structure, and concentration of the nanoparticles, as well as on the type of polymeric matrix [26-33].

Great expectations have focused on costless nanoparticles. However, it has been concerned in this paper about the effect of types of cost-fewer nanoparticles on electrical properties of polymeric nanocomposite films. With a continual progress in polymer nanocomposite films, this research depicts the effects of types and concentration of cost-fewer nanoparticles in electrical properties of industrial polymer material. Experimental results have been discussed the effects of clay and fumed silica nanoparticles with various volume fractions and temperatures on electric and dielectric properties of polypropylene.

2. EXPERIMENTAL SETUP

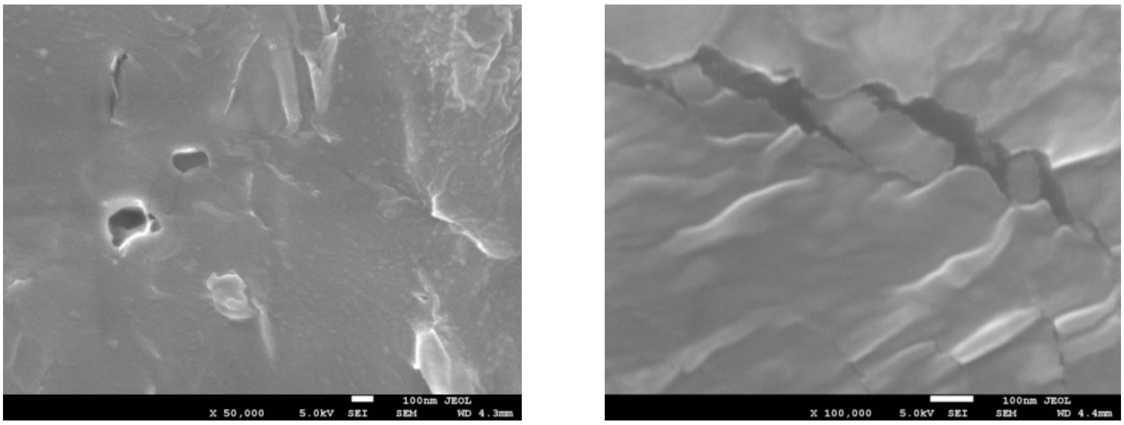
2.1. Nanoparticles

The used cost-fewer nanoparticles have a spherical particle shape (Dia.: 10nm) and have the most important characteristic for enhancing polymer applications. The reason of selection clay nanoparticles is due to having a greater effect on properties such as viscosity, stiffness and strength, using clay as nanoparticles give high levels of flame retardancy to the produced composite. Cost-fewer clay nanoparticles are catalyst to be the best filler among nanoparticles industrial materials. On the other wise, fumed silica is a fluffy white powder with an extremely low density. Also, fumed silica powder is used in paints and coatings, silicone rubber and silicone sealants, adhesives, cable compounds and gels, printing inks and toner, and plant protection.

2.2. Base Matrix

Polypropylene is one of the most common and versatile thermoplastics in the plastics industry. The properties of polypropylene have a low density, high transition temperature, high melting point, and high thermal dimensional stability. Also, it is one of the most important polymers where it is used as insulation of modern HV capacitors. Filling polypropylene with a certain nanoparticles may increase electrical, tensile & impact strength, flexural modulus, and deflection temperature properties. The final properties depend mainly on the type and percentages of nanoparticles. Generally improving tensile strength may adversely affect on corresponding elongation [10]. Polymer nanocomposite can be prepared using three main methods; intercalation [12], Sol-Gel using a hydrolysis reaction and condensation polymerization of metal alkoxide [13], directs dispersion in which inorganic nanoparticles are distributed into a matrix polymer [14]. Polypropylene nanocomposite films were prepared and composited with nanoparticles of fumed silica and montmorillonite clay (1%wt. up to10%wt.); these nanoparticles were mixed and heated up to 185 oC and 50 rpm for 8 min in kneader.

The compounded materials were ground and rolled at 160 ^{o}C to obtain thin film (thickness of 0.1±0.01) as shown in Figure 1. Polypropylene nanocomposite films were obtained under 25 MPa and 185°C for 5 min using hot press. Characterization of nanostructured material samples was carried out and achieved through a screening of the main electric and dielectric properties. The base of all nanocomposite polymer materials have been measured their electric and dielectric properties after manufacturing and detailed as shown in Table 1. Surface analysis and montmorillonite clay and fumed silica nanoparticles were examined using scanning electron microscope as shown in Figure 1.



(a) Clay/PP nanocomposite (b) Fumed silica/PP nanocomposite

Figure 1. SEM images for PP nanocomposite films

Table 1. Dielectric constant and resistivity properties of nanocomposite films

Materials	Dielectric Constant at 1kHz	Resistivity (Ω.m)
Pure PP	2.28	10^8
PP + 1%wt Clay	2.21	10^9
PP + 5%wt Clay	1.97	10^9-10^{10}
PP + 10%wt Clay	1.75	10^{10}-10^{12}
PP + 1%wt Fumed Silica	2.29	10^7
PP + 5%wt Fumed Silica	2.37	10^7-10^5
PP + 10%wt Fumed Silica	2.47	10^5-10^4

2.3. Measurement Devices

Figure 2 shows HIOKI 3522-50 LCR that measured electrical parameters of nano-metric solid dielectric insulation specimens at various frequencies: |Z|, |Y|, θ, Rp (DCR), Rs (ESR, DCR), G, X, B, Cp, Cs, Lp, Ls, D (tan δ), and Q.

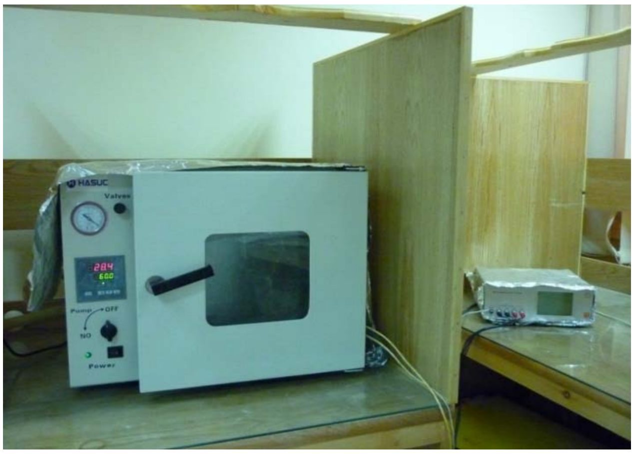


Figure 2. HIOKI 3522-50 LCR Hi-tester device

Specification of LCR is Power supply: 100, 120, 220 or 240 V(±10%) AC (selectable), 50/60 Hz, and Frequency: DC, 1 mHz to 100 kHz, Display Screen: LCD with backlight / 99999 (full 5 digits). Voltage Rating: 220V AC 50 Hz, Test Voltage: 0~5KV, 0~12KV. Output Capacity: up to 1KVA, Cut-off current: Adjustable levels current, Alarm System: Buzzer & N-GO indicator lamp. Test Time: approx 0~180 sec (Adjustable), Circuit Protect: Circuit Breaker.

2.4. Physical and Mechanical Properties of Polypropylene (PP)

Polypropylene (PP) is one of the most common and versatile thermoplastics in the plastics industry. The properties of polypropylene have a low density, high transition temperature, high melting point, and high thermal dimensional stability. Also, it is one of the most important polymers where it is used as insulation of modern HV capacitors. Filling polypropylene with a certain nanoparticles may increase electrical, tensile & impact strength, flexural modulus, and deflection temperature properties. The final properties depend mainly on the type and percentages of nanofillers. Generally, improving tensile strength may adversely affect on corresponding elongation. Physical and mechanical properties of polyproplene are listed in Table (2) [34-37]. It is found that trapping properties of matrix are highly modified by the presence of costless nanofillers clay. The nanoclay particles were dispersed homogenously in PP up to 10%wt/wt. Dielectric constant of PP-nanoclay composites was decreased successfully from 2.28 to 1.75 at 1KHz and 10% concentration. A small addition of nanoclay to polyproplene showed appreciable improvement in the electric resistivity at different frequency.

Table 2. Physical and Mechanical Properties of Polypropylene (PP)

Properties	PPs
Density, g/cm³	0,95
Yield stress, MPa	32
Elongation at yield, %	8
Elongation at break, %	70
Tensile modulus of elasticity, MPa	1300
Notched impact strength, kJ/m²	6
Ball indentation hardness, MPa	70
Shore hardness (D)	72
Mean coefficient of linear thermal expansion,	1,6x10-4
Thermal conductivity, W/m * K	0,22
Dielectric strength, kV/mm	22
Surface resistivity, Ohm	1014
Temperature range, °C	to +100

In this study, polypropylene nanocomposites were processed and characterized. Materials selection and electrical/mechanical predictable models were carried out using Cambridge Engineering Selector (CES) program. It was found that nanosilica has an adverse influence on dielectric properties of polyproplene nanocomposites while caly nanoparticles improved the electrical insulation properties. Therefore, Sol-Gel technique was applied to prepare polyproplenenanocomposites. Surface analysis and montmorillonite clay nanoparticles dispersability were examined using scanning electron microscope. Dielectrical properties were assessed using HIOKI 3522-50 LCR Hi-tester device. An experimental work for conductance and suscptance of the new nanocomposite materials have been investigated and compared with unfilled industrial materials in a frequency range of 1Hz –0.1MHz. The resulting values fits with Lichtenecker's equation [10]:

$$log\varepsilon_{com} = (1-f)log\varepsilon_m + flog\varepsilon_f \quad (1)$$

$$log\varepsilon_{com} = log\varepsilon_{com} + f(1-k)log(\frac{\varepsilon_f}{\varepsilon_m}) \quad (2)$$

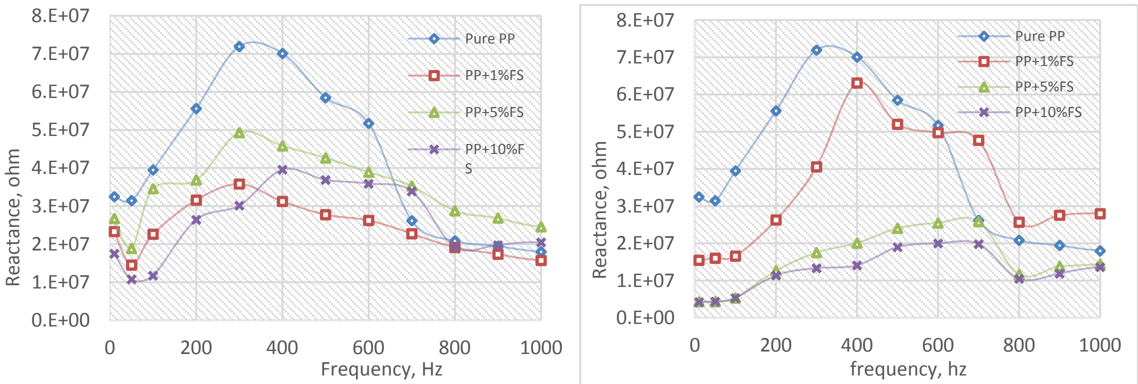
Where, f is the volume fraction of Polymer and nanoparticles. ε_m and ε_f are the relative permittivities of pure polymer and nanoparticles. k is a fitting factor of the nanocomposite.

3. RESULTS AND DISCUSSION

Beginning with a complete analysis of the dielectric spectroscopy results, this paper presents in detail the experimental results used in such specialized techniques as high-frequency dielectric measurements under variant thermally conditions. This research covers electric properties of new polypropylene nanocomposite films with new applications in a range of polymeric systems. The conductance and reactance were measured as a function of frequency in the range up to 1 kHz at variant temperatures for all samples.

3.1. Electrical Characterization of Polypropylene Nanocomposite Films at Temperature 20^{o}C

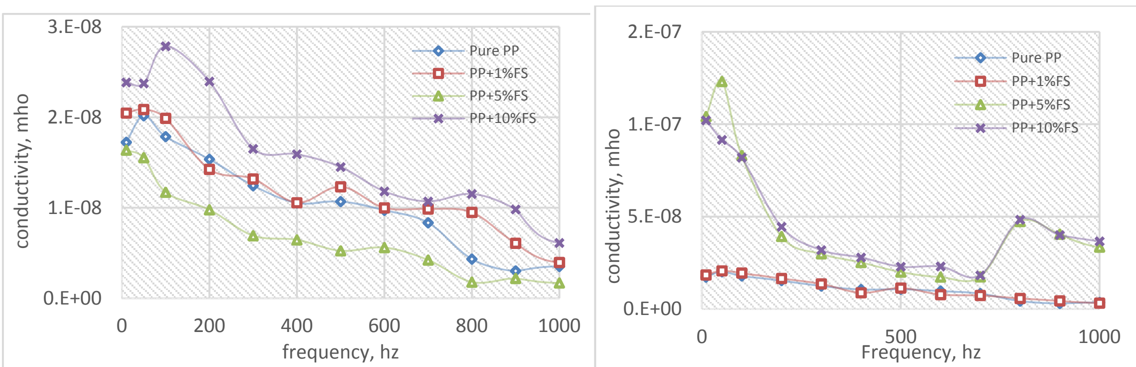
Figure 3(a) shows reactance of the tested samples as a function of frequency for clay/PP nanocomposite films at room temperature (20^{o}C). The measured reactance contrasts on decreasing the reactance with increasing the percentage of clay nanoparticles up to 1%wt; then it is increasing with increasing clay nanoparticles percentage up to 10%wt, specially, at high frequencies. Whatever, Figure 3(b) contrasts on the measured reactance that decreases with increasing percentage of fumed silica nanoparticles in the nanocomposite up to 10wt%.



(a) clay/PP (b) fumed silica/PP

Figure 3. Measured reactance of nanocomposite films at room temperature (20^{o}C)

Figure 4(a) contrast on conductance of clay/PP nanocomposite samples that decreases with increasing frequency at room temperature (20^{o}C). On the otherwise, Figure 4 obvious the measured conductance that decreases with increasing percentage of clay nanoparticles in the nanocomposite up to 5%wt but the measured conductance of clay/PP nanocomposite films increases with increasing the clay percentage of nanoparticles up to percentage of 10%wt. Whatever, Figure 4(b) shows that the measured conductance are increased with increasing the percentage of fumed silica nanoparticles in the nanocomposite especially at low frequencies. It is cleared that nanoparticles have been changed electric polymer properties. The dielectric properties of insulating polymer nanocomposite films have been investigated in the frequency domain from 0.1Hz to 1kHz.

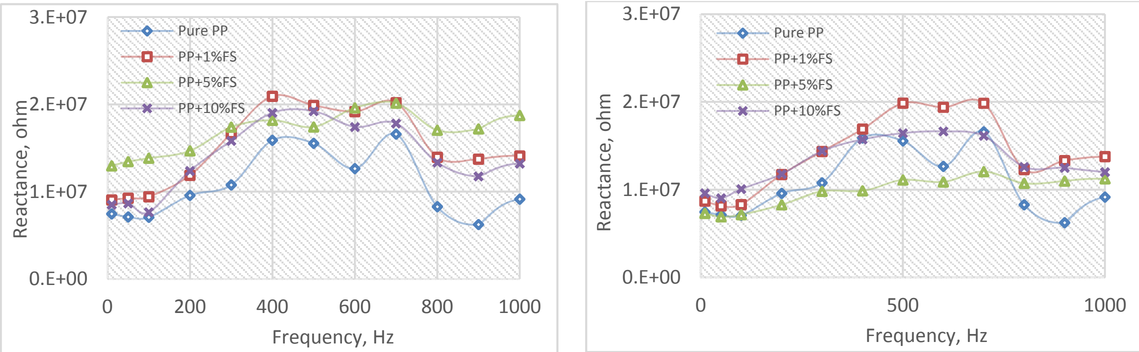


(a) clay/PP (b) fumed silica/PP

Figure 4. Measured conductance of nanocomposite films at room temperature (20^{o}C)

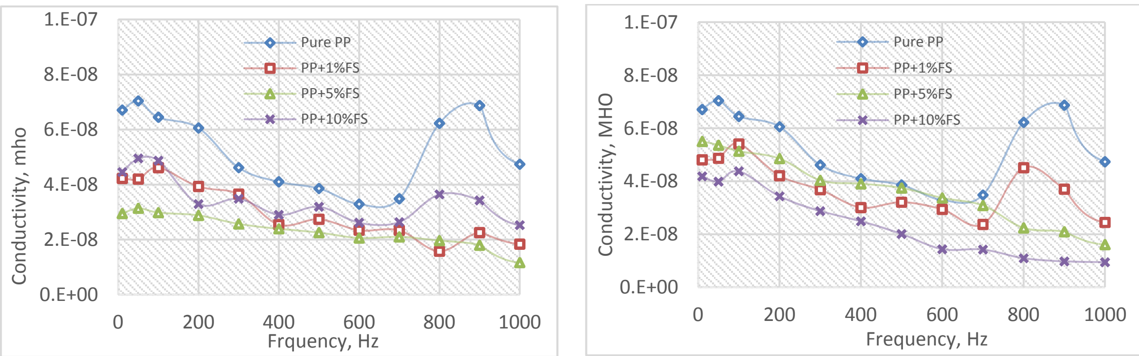
3.2. Electrical Characterization of Polypropylene Nanocomposite Films at Temperature 60°C

It is important to study thermal stability of the new polypropylene nanocomposite films in order to apply and use in industrial applications safely; it has been restricted related to the lowest melting point of both nanoparticles and polymer matrix. Thus, Figure 5(a) shows the relation between reactance versus the applied frequency for clay/PP nanocomposite films at temperature (60°C). The measured reactance of polypropylene nanocomposite increases with increasing clay percentage nanoparticles up to 5wt%. After that, the reactance of clay/PP nanocomposite decreases with increasing clay percentage nanoparticles up to 10wt%. Also, Figure 5(b) shows that the measured reactance of fumed silica/Polypropylene decreases with increasing fumed silica nanoparticles percentage up to 10%wt. Figure 6(a) depicts conductance versus the applied voltage frequency for clay/PP nanocomposite films at temperature (60°C), the measured conductance decreases with increasing clay nanoparticles percentage up to 5%wt, then, the measured conductance increases with increasing clay percentage nanoparticles up to 10%wt. Although, Figure 6(b) shows that the measured conductance decreases with increasing fumed silica percentage nanoparticles up to 1%wt, after that, the conductance of fumed silica/PP nanocomposite films decreases with increasing fumed silica nanoparticles percentage (5%wt -10%wt). This is obvious that, rising temperature of nanocomposite materials raises nanoparticles temperatures which changing dielectric behavior with respect to normal conditions.



(a) clay/PP (b) fumed silica/PP

Figure 5. Measured reactance of nanocomposite films at certain temperature (60°C)



(a) clay/PP (b) fumed silica/PP

Figure 6. Measured conductance of nanocomposite films at certain temperature (60°C)

3.3. Trends of Cost-fewer Nanoparticles on Electrical Characterization of Polypropylene

In the beginning, with comparing results for depicting the effect of raising concentration of clay and fumed silica nanoparticles are pointed out in Figures (3-4) at room temperature (20°C), the measured reactance characteristics varies between low and high with respect to percentages of nanoparticles and exposed power frequencies. On the otherwise, the measured conductance varies with increasing percentage of clay nanoparticles inside the nanocomposite, whatever, the measured conductance are closed with increasing the percentage of fumed silica nanoparticles in the nanocomposite. With rising samples temperature up to 60oC, it can be noticed that the effects high temperature values on nanoparticles inside the nanocomposite films and so, the effect of raising concentration of nanoparticles is pointed out in Figures (5-6). It is obvious that rising temperature of nanocomposite materials affects on nanoparticles heating temperatures which changing electrical behavior over the normal conditions; thus, the measured reactance has been changed with respect to weight percentages of clay or fumed silica nanoparticles up to 10%wt. Finally, the importance of adding nanoparticles of clay or fumed silica can be concluded in controlling in increasing or decreasing the electrical strength of pure polypropylene by using nanotechnology techniques. Also, increasing environment temperature of nanocomposite materials causes nanoparticles heating temperatures that changing electrical behavior over the normal conditions.

3.4 Predictable Mechanical and Electrical Behaviour of Fumed Silica/Polypropylene Composites

CES-Edu software was used to predict the electrical and mechanical properties of polypropylene nanocomposite films. Selected materials were chemically treated for polymer composites synthesis. Polypropylene nanocomposite films were electrically analyzed using LCR Hi-tester tester. Surface analysis of nanocomposite films was carried out using SEM test. The essential steps to be adopted for this research have been mentioned; thus, mechanical, physical and chemical properties of applied materials were generated. Figure 7 illustrates the electrical and mechanical properties of fumed silica/PP using CES software. Addition of fumed silica to polypropylene leads to increase dielectric constant and conductance. The initial results for using the predictable model (CES-Software) showed that addition of fumed silica to polypropylene nanocomposite films can improve slightly the tensile strength of polypropylene nanocomposite films but it causes a reduction in electrical resistivity. Therefore the fumed silica was excluded in the experimental work for cable and electrical insulation applications. Technical/management challenges and obstacles were encountered during synthesis and processing of nanocomposite films. It was noticed that clay nanoparticles agglomeration can be avoided at high mixing rate at a concentration up to 10%wt.

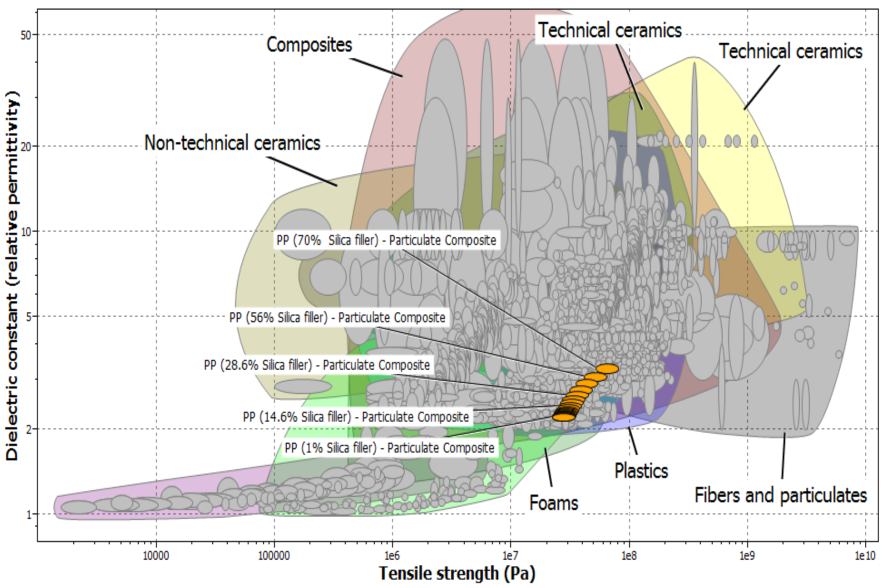


Figure 7. Electrical and mechanical properties of Fumed Silica/PP using CES software

3.5 Consequence of Nanotechnology Science on Dielectrics Properties

In the field of dielectrics and electrical insulation, polymer nanocomposites are known for their excellent dielectric properties and are the subject of intensive research. For example, polymer-clay nanocomposites are considered promising as electrical insulation for power apparatus, cablesand wires in the near future due to their good insulating capabilities, flame retardance and mechanical properties [34]. Thermoplastic and thermoset resins reinforced by nanoparticles of clay, silica, rutile and alumina have been developed and studied [35]. It has been synthesized well-dispersed polypropylene based nanocomposite dielectric materials via in situ supported metallocene polymerization catalysis, to investigate the effects of matrix polymer and nanoparticle identities, loading, and shape on the electrical/dielectric properties of the resulting nanocomposites, and to compare the experimental findings with theoretical calculations. The investigation of electrical properties of cenosphere filled polypropylene composites informs that Dielectric constant decreases with increasing cenosphere volume fraction. It is found to decrease with test frequency and increased with temperature. Dissipation factor follows the trend observed for dielectric constant with respect to the material and electrical test parameters. Both a.c. and d.c. conductivity have a strong dependence on the weight percent of fly ash in polypropylene. Validation of theoretical and experimental dc conductivity values shows satisfactory results at higher concentration of cenosphere [36]. New types of coupling agents were synthesized and applied for enhancing the interaction between talc supported multi-walled carbon nanotubes and polyproplene. Carbon nanotubes can lower the electrical resistivity of polypropylene considerably. The electrical conductivity percolation threshold in the polymer matrix is approx. 2 wt% for both nanotube types, the PP/CNT composites were investigated on the basis of the measured data; carbon nanotubes exhibited very similar behaviour as polypropylene fillers. The thermal conductivity of polypropylene can be improved by adding carbon nanotubes into the matrix. UPNT nanotubes outperformed commercial NC nanotubes by 17% in this respect. Unlike in the case of electrical conductivity, thermal conductivity does not saturate at approx. 2 wt% CNT concentrations but rather, it keeps increasing as a quasi linear function of the nanotube contents of the sample [37].

4. CONCLUSION

As the electrical insulation of polypropylene composites contribution to its reactance and conductance value in polypropylene nanocomposite films in lower frequency range may result in the electrical insulation of the nanocomposite films having been affected by the presence of nanoparticles. There is no linear performance insight of increasing percentage of clay nanoparticles on polypropylene electrical properties. Whatever, increasing percentage of fumed silica nanoparticles increases polypropylene conductance but decreases its reactance at high thermal conditions. Thermal stability of the new polypropylene nanocomposite films has been restricted related to the lowest melting point of both nanoparticles and polymer matrix for getting industrial safely applications. Under high thermal conditions, the influence of the relaxation time of the charge carriers on the electrical insulation of polypropylene nanocomposite films can be ignored. Thus the number of charge carriers and applied frequency become dominating factors of the electrical insulation of polypropylene nanocomposite films. The presence of nanoparticles inside polypropylene will restrict the chain mobility and result in increasing electric insulation as such restriction limited the generation of mobile charge and the movement of charge carriers in polymer dielectrics, especially at a lower frequency range where the insulation will play a more important role. Thus the variation of reactance and conductance value at low frequency range may be due to the influence of inorganic fillers' electrical insulation. Electrical stability of new nanocomposite films occurs at small amounts clay or fumed silica nanoparticles but adding large amounts these nanoparticles to polypropylene will be reverse electrical behavior characteristics gradually. A high thermal condition of polypropylene nanocomposite materials is changed electrical behavior over the normal conditions.

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BIOGRAPHIES OF AUTHORS

Ahmed Thabet was born in Aswan, Egypt in 1974. He received the BSc (FEE) Electrical Engineering degree in 1997 and MSc (FEE) Electrical Engineering degree in 2002 both from Faculty of Energy Engineering, Aswan, Egypt. PhD degree had been received in Electrical Engineering in 2006 from El-Minia University, Minia, Egypt. He joined with Electrical Power Engineering Group of Faculty of Energy Engineering in Aswan University as a Demonstrator at July 1999, until; he held Associate Professor Position at October 2011 up to date. His research interests lie in the areas of analysis and developing electrical engineering models and applications, investigating novel nano-technology materials via addition nano-scale particles and additives for usage in industrial branch, electromagnetic materials, electroluminescence and the relationship with electrical and thermal ageing of industrial polymers. Many of mobility's have investigated for supporting his research experience in UK, Finland, Italy, and USA …etc. On 2009, he had been a Principle Investigator of a funded project from Science and Technology development Fund "STDF" for developing industrial materials of ac and dc applications by nano-technology techniques. He has been established first Nano-Technology Research Centre in the Upper Egypt (http://www.aswan.svu.edu.eg/nano/index.htm). He has many of publications which have been published and under published in national, international journals and conferences and held in Nano-Technology Research Centre website.

Youssef A. Mobarak was born in Luxor, Egypt in 1971. He received his B.Sc. and M.Sc. degrees in Electrical Engineering from Faculty of Energy Engineering, Aswan University, Egypt, in 1997 and 2001 respectively and Ph.D. from Faculty of Engineering, Cairo University, Egypt, in 2005. He joined Electrical Engineering Department, Faculty of Energy Engineering, Aswan University as a Demonstrator, as an Assistant Lecturer, and as an Assistant Professor during the periods of 1998–2001, 2001–2005, and 2005–2009 respectively. He joined Artificial Complex Systems, Hiroshima University, Japan as a Researcher 2007–2008. Also, he joined Faculty of Engineering, King Abdulaziz University, Rabigh, Saudi Arabia as Associate Professor Position at April 2014 up to date. His research interests are power system planning, operation, optimization, and techniques applied to power systems. Also, his research interests are wind energy, and nanotechnology materials via addition nano-scale particles and additives for usage in industrial field.