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ADVANCED SIMULATION MODELS FOR PREDICTING ELECTRIC RESPONSE AND AGING OF NANOCOMPOSITE INDUSTRIAL INSULATION MATERIALS

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Electric response of nanocomposite industrial materials has been investigated for all possible causes and effects of the interphase region on a variety of complex composite systems by using Interphase Power Law IPL model. Thus, this paper investigates the effective dielectric response of nanocomposite industrial insulation materials and effects of the interphase region on a variety of complex composite systems. According to the statistical approach of failure in polymeric insulators, that leads us to predict the procedures and mechanisms of failures which occur within insulating materials, this paper illustrates the failure statistics of a life model for ac electrical aging of nanocomposite insulation industrial materials which depicts the mechanisms of electrical breakdown under high voltage electric fields by advanced simulation model program. Case studies are used to illustrate the way and mechanisms which mechanistic features which can be related to the parameters of the failure statistic. Realized lifetime of insulation materials can be evaluated by means of this model. Aging of polymeric insulators has been examined through various high voltage electric fields, so the possibility of predicting the lifetime of individual specimens has been succeeded.

KEYWORDS: *Nanocomposite, Nanofillers, Life-time, Statistical, Polymers, Electrical aging, thin films.*

1- INTRODUCTION

Nano-filler clay has created novel costless industrial materials by getting smoothly changing in the effective dielectric constant with the variation of filler volume fraction [1]. Power law model relationships are used in dielectric modeling of composite systems. The general two component power-law model for complex permittivity has been used extensively for a wide range of material systems with varied success, including air-particulate composites, ceramic-ceramic composites and polymer-ceramic composites. Research into the physical and electrical characteristics of particulate filled polymer composites has supported the existence of interphase regions at polymer-dispersant interfaces [2-7]. IPL model in its general form is commonly known as the Lichtenecker-Rother equation. However, a clear distinction exists between the IPL model and the Lichtenecker-Rother LR model. The IPL model is unique in that it incorporates two independent components of the composite as well as an interphase region that is dependent upon the characteristics of the two components.

This model thus does not rely on three independent components but rather two independent components and the interphase region between them. The LR model is based on a composite consisting of three independent constituent components with no interactions between the components. It is important to recognize the distinction between the IPL model and a standard three-component power-law mixture. The determination of the interphase thickness is further described in detail in [8-16]. The interphase overlap probability arises from the proximity of the individual filler particles. When the filler particles come close together, the interphase regions surrounding each filler particle begin to overlap, thereby reducing the effective interphase volume fraction. The interphase overlap probability is a function of the filler volume fraction, interphase thickness and shape and size of the filler particles. Approximations have been developed to estimate the overlap of such particles or alternately analytical solutions to percolation models. A general interphase overlap probability function has been developed for spherical particles. In several investigations, this interphase region has been shown to have significantly different properties compared to the bulk resin. In recent years, polymer nanocomposites have been extensively studied in optical, thermal and mechanical properties, but there has been relatively little research into dielectric properties [17-21]. Deterministic theories of electrical breakdown are very satisfying to the scientists and engineer, therefore, it would appear that it is possible to design a system that simulate the electrical breakdown in insulating materials at applied fields well below the critical values predicted for the various failure mechanisms, and with a wide distribution of times to failure. Because of this, the reliability estimates required by manufactures and users need a statistical analysis of a series breakdown tests. So far most of the mechanisms considered have been clearly defined phenomena even if the precise details whereby they cause failure are not known [22-27].

2- ANALYTICAL MODELS

2.1 Interphase Power Law IPL Model

The interphase region is comprised of polymer molecules that are bonded or otherwise oriented at the matrix-filler interface. This interfacial-bonding region is termed the interphase and results from the confining effect that the rigid filler particles have on the mobility of the polymer molecules in the matrix. In the bulk matrix regions of the composite, the polymer chains adopt random orientational configurations. IPL model is based on a simple extension of a general power law model in which a composite system containing filler, interphase and matrix regions may be treated as a unique three-component composite system comprising two primary components matrix and filler, and an inter-phase region that is inextricably dependent upon the characteristics of the filler and matrix components [10]. Power law relationships are quite often used in dielectric modeling of composite systems. More generally, for a composite comprised of n number of components, the power law mixtures model may be written as:

$$\varepsilon_c^{\beta} = \sum_{i=1}^{n} \varphi_i \varepsilon_i^{\beta} \qquad (1)$$

Where, ε_c and ε_i are the complex permittivity ($\varepsilon'+j\varepsilon''$) of the composite system and any constituent component of the composite respectively. Also, ε' is the effective permittivity of the materials. While, φ_i is the volume fraction of the constituent component, and β is the filler particle shape and orientation. The n-component power-law relationship described by equation (1) is easily extended to a three component composite system, as described by [7]:

$$\varepsilon_c^{\beta} = \varphi_f \varepsilon_f^{\beta} + \varphi_i \varepsilon_i^{\beta} + \varphi_m \varepsilon_m^{\beta} \quad (2)$$

For a composite model that contains an interphase region, φ_f, φ_i, and φ_m represent the volume fraction of the filler component, the interphase region and the matrix component of the composite, respectively, ε_c, ε_f, ε_i and ε_m are the complex permittivities of the composite, the filler, interphase and matrix regions of the composite, respectively. It is important to recognize the distinction between the IPL model and a standard three-component power-law mixture. Therefore, refer to independent composite constituents as components and the dependent constituents as interphase. The distinction between these cases will become more evident with further explanation. The filler volume fraction of equation (2), φ_f is directly measured for a given composite system. The matrix volume fraction is given by $\varphi_m = (1 - \varphi_f - \varphi_i)$. The interphase region volume fraction, φ_i, is dependent upon the filler volume fraction, the filler surface area and the thickness of the interphase region surrounding each filler particle.

The interphase volume fraction φ_i, is determined for monodisperse, spherical particles by [8]:

$$\varphi_i = \frac{\frac{4\pi\varphi_f}{3}\left[\left((r+\Delta r)^3 - r^3\right) - F\,6\left(3(r+\Delta r) - \Delta r\right)\Delta r^2\right]}{r^3} \quad (3)$$

Where r is the radius of the filler particles, Δr is the thickness of the interphase region and F is an overlap probability function. Assuming that, there are no areas of interphase overlap that would reduce the total volume of interphase. Therefore, the interphase volume fraction, φi, is calculated by:

$$\varphi_i = (1 - F)(S_f . \Delta r).\rho_f . \varphi_f \quad (4)$$

Where S_f is the specific surface area of filler (measured in m2/g), and ρf is the density of filler (measured in g/m3) [5].

The interphase density is assumed to be equal to that of the matrix component since it is comprised of matrix molecules. Typically, the interphase thickness is on the order of one molecular radius of gyration (5 to 20) nm. The interphase overlap probability F arises from the proximity of the individual filler particles. The interphase overlap probability is a function of the filler volume fraction, interphase thickness and the shape and size of the filler particles. Approximations have been developed to estimate the overlap of such particles using analytical solutions to percolation models [9-12]. For spherical inclusions and for randomly oriented ellipsoids, Landau and Lifshitz as well as Looyenga determined that β=0.33. A general interpretation of β based on the inclusion shape specifically ellipsoid axes lengths. Equations (2) through (4) can be used to complete a generalized, comprehensive model for the prediction of the complex dielectric permittivity of composite systems containing interphase bonding regions [10].

This model discusses a simple interphase analytical model which is capable of providing additional insight to both the real and imaginary parts of the complex permittivity of composite systems incorporating the interphase theory. The composite system containing filler, interphase and matrix regions is illustrated in Fig.1. A composite system comprising two primary components (matrix and filler) and an interphase region may be treated as a unique three-component composite system in which the interphase volume is inextricably dependent upon the characteristics of the filler component.

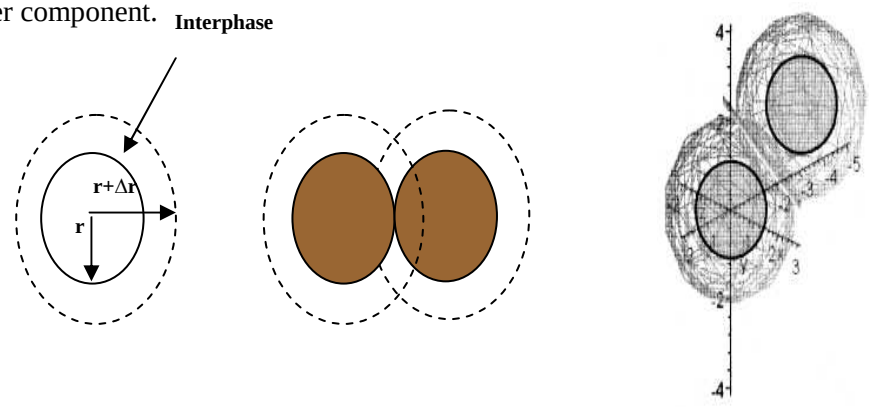

Fig.1 Interphase region surrounding the filler particles in a composite system

2.2 Lifetime Breakdown Model

The contending life expressions all are related to the characteristic time to failure. It is therefore interesting to know the failure statistics. Methodology of the lifetime model simulates any dielectric material as a grid as shown in figure (2), it shows the structural bonds of the insulation material. In this model, black points are the boundaries of the dielectric material whatever; the top electrode is assigned with a high voltage while the bottom electrode is assigned with zero volts [25]. The distance between any adjacent points on the grid are to be considered as capacitors and so, the boundaries are fixed at uniform field and cannot be failed.

Each bond in the model is assigned a value for a specific property; these values will be a set of randomly selected numbers. Thus, the initial field in each bond is as follows:

$$E_n = \frac{V}{L/N} \qquad (5)$$

Where,

E: is the electric field in each bond (kV/mm)
V: is the voltage applied on the insulation material (kV)
L: is the thickness of the insulation material (mm)
N: is the total number of bonds within the thickness of the insulation material
n: is the bond order in the array

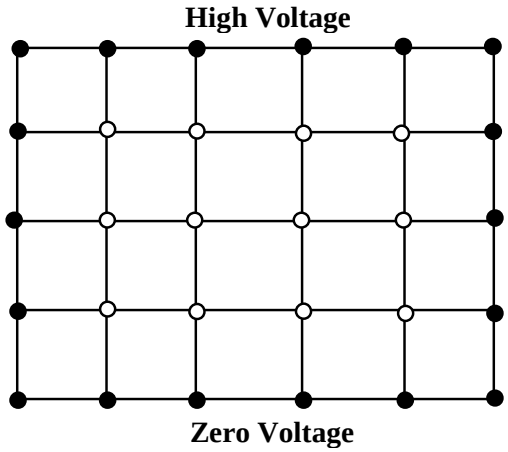

Fig.2 Lattice diagram of the structural bonds of the insulation material

The shortest time left before failure of local bonds has been calculated as follows [25]:

$$t_{n+1} = \frac{h}{T}\frac{3.8672 \text{ x } 10^{22} \exp\left(\frac{17762 - 0.791\,E_n^{1.556}}{T}\right)\ln\left(\frac{A_{eq}(E) - A(t)}{A_{eq}(E) - A^*}\right)}{Cosh\left(\frac{153 - 0.791\,E_n^{1.556}}{T}\right)} \quad (6)$$

Where,

T: is the absolute temperature in K

h: is the planck constant. Its value is 6.626068 x 10-34 (m^2kg/s)

A^*: is the property which is given the randomly distributed value on the bonds in the grid, and so, its value is changed from 0.01 to 0.99 with the average value at 0.485[5].

Aeq (E): is the equilibrium value of the same quantity of A*, and it can be estimated as follows:

$$A_{eq}(E) = \frac{1}{1 + \exp\left(\frac{306 - 1.582\,E_n^{1.556}}{T}\right)} \quad (6.1)$$

A(t): is the measure of degradation remaining within the bonds at a time t seconds, this means that the bond has not failed but the others of grid-bonds are failing, so it can be evaluated as follows:

$$A(t) = A_{eq}(E_n).\left(1 - \sum_{n=1}^{n+1}\exp\left(D(E)\,t_n\right)\right) \quad (6.2)$$

$$D(E) = 2.58585\text{x}10^{-23}\frac{T}{h}\exp\left(\frac{-17762 + 0.791E_n^{1.556}}{T}\right)Cosh\left(\frac{153 - 0.791E_n^{1.556}}{T}\right) \quad (6.3)$$

The breakdown of a bond will occur when the degradation, A(t) is existing in the bond at a time t which is equal to the property that is randomly distributed on the bond in the grid, i.e. A^*. However, as the first bond fails, this may cause the adjoining bonds to fail next as the potential across the adjacent bonds may increase. This will mean recalculating the field in each bond once a bond fails, and scanning the bonds for the shortest time left before failure. This time will be added to the counter and a plot of the degraded structure and time will be illustrated.

3- APPLICATIONS AND SELECTED NANO-FILLERS AND POLYMERS FOR INDUSTRIAL MATERIALS

3.1 Selected Nano-Fillers Industrial Materials

Clay mineral used in the polymer industry is kaolinite. Kaolinite is the main constituent of filler clays, significant quantities of other minerals can often be present. The particle shape is the most important characteristic of kaolinite for polymer applications. The platy nature means that clay fillers have a greater effect on properties such as viscosity, stiffness and strength, using clay as nano filler gives high levels of flame retardancy to the produced composite, and it's selected in this study. Clay is considered a catalyst to be the best filler among nano-fillers industrial materials and Costless.

3.2 Selected Polymers for Industrial Materials

Many selected polymers industrial materials are used in this paper, such as Acrylonitrile Butadiene-Styrene ABS has been created with rubber particles in order to increase the toughness. ABS has good electrical properties that are fairly constant over a wide range of frequencies. Polyethylene PE applications vary depending upon the grade of resin LDPE, because of its flexibility, is used primarily for prosthetic devices and vacuum formed parts. Polyvinyl Chloride PVC is the most widely used of any of the thermoplasts, it is stronger and more rigid than other general purpose thermoplastic materials. The primary applications for PVC include electrical conduit, and wire insulation. Finally, applications for Epoxy-based materials are extensive and include coatings, adhesives and composite materials such as those using carbon fiber and fiberglass reinforcements. Epoxy resin formulations are important in the electronics industry, and employed excellent electrical insulators and protect electrical components from short circuiting, dust and moisture. In this paper, lifetime model can be applied on the industrial insulation materials at T=383 K, under 20kV, 40kV, and 80kV. Thus, it can be shown the effect of nanofillers in the industrial polymers and failure shape mechanisms related to the deterministic and statistics of electrical breakdown failures in the new nanocomposite material by adding 10wt% clay to 90wt% PE "Specimen no.1", 10wt% clay to 90wt% PVC "Specimen no.2", and 10wt% clay to 90wt% Epoxy "Specimen no.3". Also, it has been clarified that the effects of various levels of high voltage electric fields on the deterministic and statistics of electrical breakdown failures. A number of case studies are used to illustrate the way in which mechanistic features can be related to the parameters of the failure statistics. Some selected examples will be presented in order to illustrate the methodology. Then,

the way in which knowledge of the failure mechanism can guide the interpretation and use of failure statistics will be explored. Finally, the possibility of predicting the lifetime of individual specimens will be examined briefly.

3.3 Nano-Composite Industrial Materials

An interphase layer is usually present in a composite system between the matrix and reinforcement due to sizing, chemical reaction, and diffusion process. This interphase layer modifies the stress transfer mechanism between the matrix and fiber, and could be tailored to an optimal compromise between the strength and toughness of the composite. The use of polymers in multi-component systems, such as adhesives and composites, requires sensitive, nano-scale property evaluation of polymer systems. Nano-scale properties that control various aspects of material performance can be different from bulk properties. For example, the behavior of polymer composites is highly dependent on the interfacial strength between the fiber and matrix. However, interfacial strength is controlled by the matrix material adjacent to the fiber, referred to as the fiber-matrix interphase region. This paper discussed the effect of filler volume fraction on effective dielectric constant for selected above polymers with clay selected nano-filler as a nanocomposites industrial materials which will be new costless industrial materials.

4- RESULTS AND DISCUSSION

This paper has been discussed the effects of filler permittivity; interphase permittivity, filler surface area, filler interphase thickness, and filler particle shape. The effective dielectric constant of the nanocomposite system is investigated in this research for Clay nano-fillers with four industrial polymers such as PE, Epoxy, and PVC. The composite permittivity depends on the filler, matrix, and interphase dielectric constant, which is varied from the value of the matrix toward the filler in case of insulating materials. When the filler volume fraction increases the composite permittivity decreases to a certain value depending upon polymer matrix permittivity. Table 1 depicts the properties of the industrial insulation materials characteristics for the composite systems. From these simulated results the clay shows the best behavior as filler used for insulation materials.

Table 1. Industrial insulation materials characteristics for the composite systems

Dielectric Constant Data			
Fillers	***Matrix***	***Interphase***	***Specimens***
Clay 2.0	*Epoxy 5.0*	*Clay-PE 1.099*	*2 : 2.3 Clay-PE "Specimen_1"*
Volume Fraction 0 to 1	*PE 2.3*	*Clay-PVC 1.3353*	*2 : 3.3 Clay-PVC "Specimen_2"*
Surface Area 106 m2/g	*PVC 3.3*	*Clay-Epoxy 1.6708*	*2 : 5 Clay-Epoxy "Specimen_3"*
Density 2.33g/cc		*Thickness 11nm*	
β (Spherical) 0.33			

4.1 Effect of Filler Volume Fraction

The interphase power-law model predicts a distinct non-linearity in the effective dielectric constant as a function of filler volume loading. As the volume fraction of

clay increased the dielectric constant for every polymer decreased as shown in the Fig. 3. For a filler volume fraction above 56%, the nano-composite of clay with all polymers are the same dielectric constant, while a volume fraction below 56%, the dielectric constant is lower values of clay with PE nano-composite and higher values of clay with epoxy.

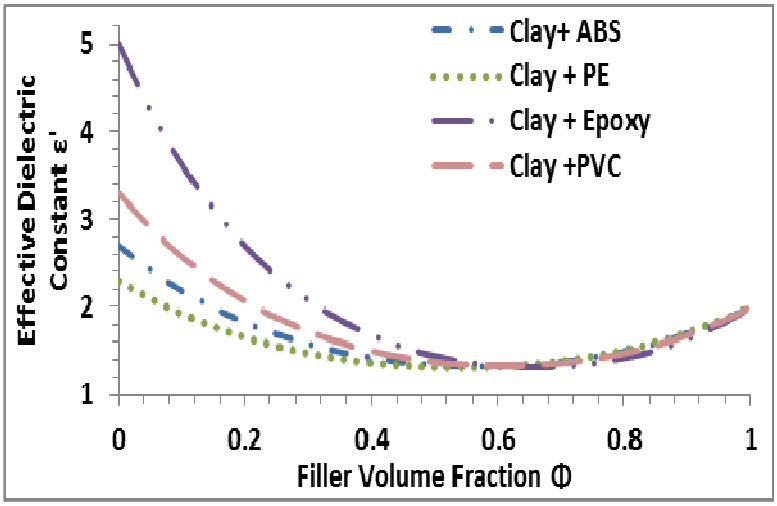

Fig. 3 Effect of filler volume fraction on effective dielectric constant for different nanocomposites industrial materials

4.2 Deterministic and Statistics for Electrical Aging Breakdown at 20KV

For many applications at 20kV, it has been evaluated that the damage to short circuit in the insulation materials and represented the structure failure which have occurred in the specimens of insulation materials. Fig. (4) illustrates that the evaluation of damage to short circuit in a random selected specimens at 20kV from the simulation lifetime program. Related to the fluctuations are governed by random failures in the specimen bonds and deterministic mechanisms, the failure shape in the specimens are simple and there are low numbers of bonds which had been failed through specimen according to applying 20kV.

The damage occurs at the weakest bonds in various positions in the specimens with respect to the electric fields and potentials which applied on each bond. The failure mechanisms in the specimens have been shown in Fig. (5) and so it has been taken various mechanisms according to the nature of random weakest bonds, and it has been started from the weakest bonds in any way of the specimens and extended gradually in any direction (up, down, left, and right) within the specimens according to the electric fields equilibrium which affects on each bond and so, the number of failed bonds has been changed with respect to time from specimen to another according to the nature of the bonds in specimen.

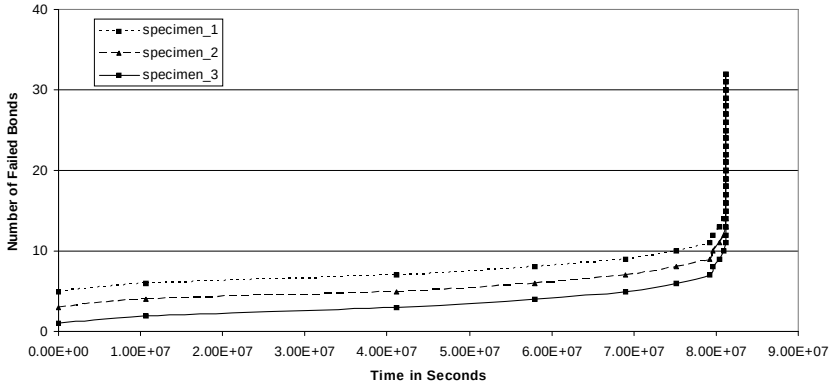

Fig. 4 Damage to short circuit in insulation material specimens at 20kV

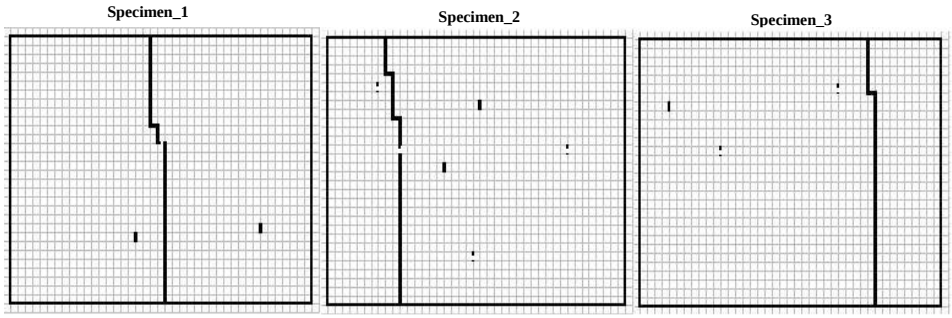

Fig. (5): Structural failure mechanisms in insulation material specimens at 20kV

4.3 Deterministic and Statistics for Electrical Aging Breakdown at 40KV

With respect to many failure specimens at 40kV, figure (6) illustrates that the evaluation of damage to short circuit in various specimens at 40kV by the author. So, the damage occurs at the weakest bonds in different positions in the specimen with respect to the electric fields and potentials which applied on each bond and equilibrium fields. Also, the failure mechanisms have been shown in fig. (7) didn't take one shape of failure mechanism (from top to bottom) as recognize later but it has been taken various mechanisms according to the nature of the bonds in the insulation material, therefore, it was started from the weakest bond in any position in the specimen and extended gradually in any trend according to the electric fields equilibrium which affects on each bond. Thus, the failure mechanism is like a chain which consists of two or more horizontal and vertical chains which combined together at different times. It is noticed that the number of failed bonds is increased directly with respect to time and it differs from specimen to another according to the shape of the mechanism and length of the failure. Also, the number of failed bonds is increased slowly in the beginning of applied potential then; it is increased rapidly (after forming chains) with respect to time. Also, it is noticed that, the failure shape in the specimens are not simple and the numbers of bonds which had been failed through specimens have been increased according to applying 40kV.

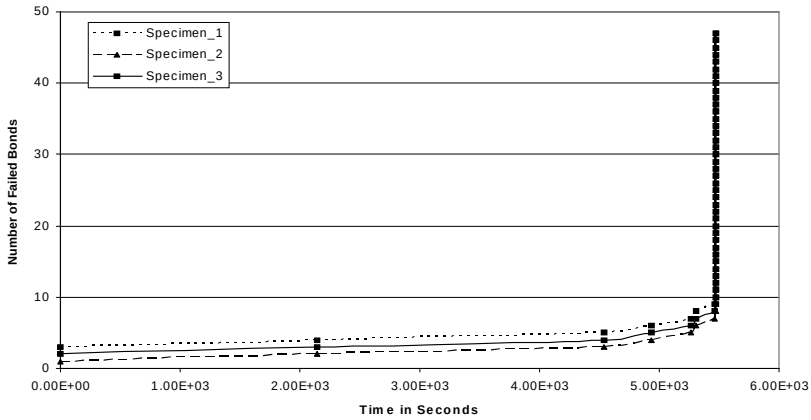

Fig. (6): Damage to short circuit in insulation material specimens at 40kV

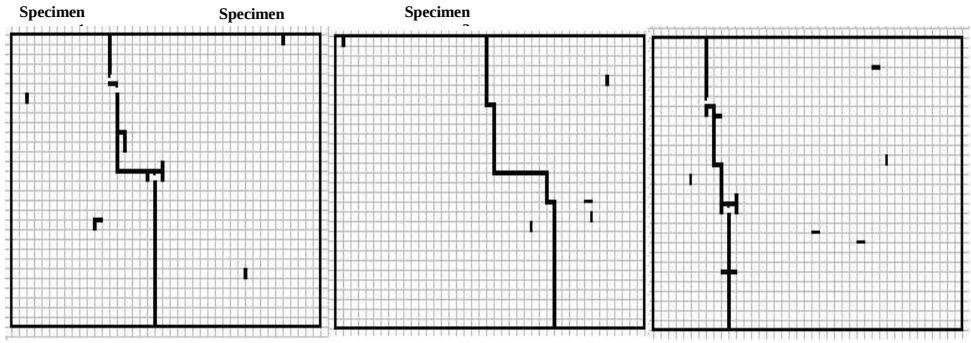

Fig. (7): Structural failure mechanisms in insulation material specimens at 40kV

4.4 Deterministic and Statistics for Electrical Aging Breakdown at 80KV

At the potential voltage 80kV, the results of failure breakdown voltage have been shown in fig. (8). All results at 80kV illustrate that the damage occurs at the weakest bonds in different positions in the specimens with respect to the electric fields and potentials which applied on each bond. It is noticed that the time of damage to short circuit in insulation material specimens decreases by increasing the applied voltage over the specimens up to 80kV. But, the numbers of failed bonds in the specimens increases with increasing the applied voltage over the specimens up to 80kV. And so, the failure shapes in the specimens are complex and the numbers of bonds which had been failed through specimens have been increased according to applying 80kV.

Also, the failure mechanisms have been shown in fig. (9) didn't take one shape (from top to bottom) as recognize later but it was taken a random shape mechanism, therefore, it has been started from the weakest bond in any position in the specimen and extended gradually in all trends (horizontal or vertical) according to the electric fields equilibrium which affects on each bond. The final failure mechanism is like a chain which can be consists of different two or more horizontal and vertical chains

which combined together at different times. It is noticed that the number of failed bonds is changed directly with respect to time and the number of failed bonds differs from specimen to another according to mechanism and length of the failure. Also, the number of failed bonds is increased slowly in the beginning of applied potential then; it is increased rapidly (after forming chains) with respect to time.

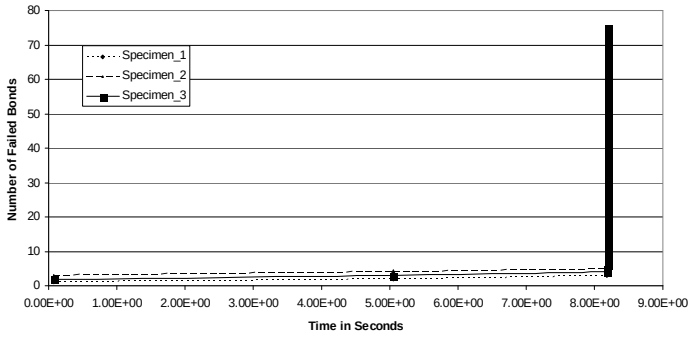

Fig. (8): Damage to short circuit in insulation material specimens at 80kV

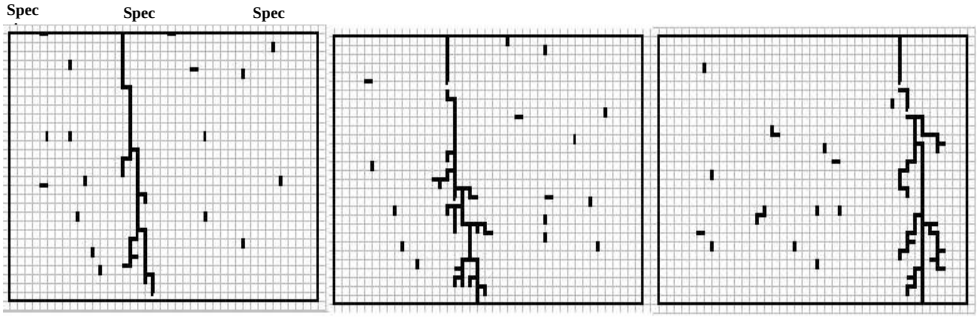

Fig. (9): Structural failure mechanisms in insulation material specimens at 80kV

4.5 Comparison Between Deterministic and Statistics

Aging process differs with respect to random defects and its electric fields and potentials; therefore, figures (4-9) illustrate the damage in insulation materials and structural failure mechanisms at various potentials 20kV, 40kV, and 80kV. Therefore, at each specimen, various damages and mechanisms occur at various positions with respect to the applied voltage. The weakest bonds in the insulation materials and its types (horizontal or vertical) are not in fixed points or positions in the insulation materials but these points have been changed with respect to the applied voltage and the nature arrangement of the weakest bonds. So, in the same insulation material specimens, the failure mechanisms have been represented in various shapes according to the equilibrium applied potentials within all bonds in the specimen. Also, it can be cleared that the numbers of failed bonds have been changed with respect to applied voltage. Also, it is noticed that the time of damage to short circuit in insulation material specimens decreases by increasing the applied voltage over the specimens, it is normally, but, the treeing failed bonds in the specimens defers related to the types of nanoparticles in the composite and the applied voltage over the specimens.

5- CONCLUSIONS

Clay has a very good record behavior as a nano-filler used for insulation materials and has an effective dielectric characteristic for different polymer industrial materials of nanocomposite insulation materials by IPL. Clay nano-fillers have been created novel costless industrial materials for getting smoothly changing effective dielectric constant with the variation of filler volume fraction of the nanocomposite material. Increasing Clay filler surface area and its interphase thickness more smoothly decreased the effective dielectric constant of the nano-composite, and the lowest effective at 55%. The damage in nanocomposite industrial materials occurs at the weakest bonds at various positions within nanocomposite industrial insulation materials with respect to the electric fields and potentials which applied on each bond. The failure mechanisms are started from the weakest bond in any position in the insulation material and extended gradually in various trends according to the electric fields equilibrium which affects on each bond therefore; the final failure mechanisms represented as various a chains which consist of different two or more horizontal and vertical chains which combined together at various times. Thus, the failure mechanism didn't take one shape (from top to bottom) but the failure mechanism and the numbers of failed bonds have been changed with respect to time and applied potential specially; the numbers of failed bonds are changed directly with respect to time and inversely with the applied potential.

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نماذج متقدمة للتنبؤ بالاستجابة الكهربائية للمواد العازلة الصناعية لمركبات النانو وعمرها الكهربي وتقادمها

نماذج المحاكاة المتطورة على التنبؤ استجابة الكهربائية وشيخوخة بمركب متناهي في الصغر مواد العزل الصناعية

ان المستوي الهيكلي الكميائي للبوليمرات المركبة والتي تشمل منطقة التداخل تحتوي علي جزيئات من مصفوفة البوليمر والتي إرتبطت بسطح حبيبات النانو المضافة للمركب تحت مسمي "حيز التداخل" والتي تحدث نتيجة لإعاقة هذه الحبيبات لحركة جزيئات البوليمر داخل المصفوفة إعتمادا علي سماحية كل من البوليمر وحبيبات النانو ومجال التداخل ونسب كل منهما داخل المركب. ومن خلال هذا البحث تم دراسة تأثير حيز التداخل علي عدد من المواد المركبة الصناعية مع تغيير نسبة الحبيبات داخل المركب. كما يلخص هذا البحث الي كيفية ابتكار مواد عازلة صناعية جديدة رخيصة التكلفة باستخدام احدث تقنيات النانوتكنولوجي. لحساب الانهيارات التى تحدث داخل المواد الكهربية العازلة فاننا نجد أن نظريةالاحصاء تقودنا الى طرق التنبؤ بانظمة الانهيارات التى تحدث فى المواد العازلة فهذة الدراسة توضح نموذج تمثيلى متقدم للتنبؤ بالعمر الكهربي للمواد العازلة المكونة من طبقات رقيقة من المواد العازلة الصناعية لمركبات النانو التى تبين تاثير انظمة الانهيار الكهربى تحت تاثير مجال الجهد العالى. فقد تم حساب عمر المواد الكهربية العازلة و زمن عمر المواد العازلة الكهربية من خلال هذة الدراسة فى مختلف المجالات و الجهود الكهربية وهكذا تم بنجاح التنبؤ بزمن عمر عينات المواد الكهربية.

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