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DC Conductivity and Tensile behavior Investigation on Fumed Silica -EPDM Nanocomposite for Electric Insulation Applications

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Abstract

Nano composites of fumed silica/EPDM have been prepared by the convenient technique, using an open mill mixer. The obtained vulcanized fumed silica/EPDM composites have been subjected to FTIR investigation to confirm the molecular structure in the composites matrix. The dc conductivity investigation was carried out for fumed silica/EPDM nano composites from room temperature up to 453 °K . The results illustrate a thermal activation with activation energy in the range 0.026 and 0.036 ev. The values of conductivity at room temperature don't show remarkable variation with fumed silica.

The stress-strain of fumed silica/EPDM nanocomposites was studied at room temperature for different concentrations of fumed silica 5 -25 phr. In addition, the tensile study for silica /EPDM illustrates yield transition for the different silica concentrations. A significant values of modulus ; resilience ; stress and strain at fracture are discussed on the basis of silica particle - silica particle ; silica - EPDM cohesions as well as composite matrix free volume .

Key words: *EPDM , Fumed silica ,Nano composite.*

1. introduction

Insulating materials made from porcelain and glasses have more than a century of service [1-8] while insulating materials made from polymers have several decades [9-15]. Nowadays the usage of polymers has been increasing every time due to the enormous advantages of polymers over porcelain and ceramics such as follows: Breakage, Ceramics are very fragile; this means that

ceramics can be broken easily in transit, handling or installation. Weight, Porcelain bodies are very massive due to the dense nature of ceramics, which lead to difficulty in handling and transportation, especially on replacement during maintenance and repairs in addition to high cost. Recently, polymer composites are used as high voltage insulation materials [9-12,14,16].

Rubbers in general have good insulation. Many authors have studied the effect of nano fillers for the reinforcement of rubbers to be used as insulating materials[17-19,21,22]. Ethylene propylene Diene Monomer (EPDM) rubber has many advantages rather than other types of rubber; environmental resistance such as UV and thermal degradation.

The present study focuses on using fumed silica as a reinforcement filler in EPDM to maintain the insulation of filled EPDM matrix. The study deals with the effect of temperature on the insulation of silica/EPDM with different concentrations of fumed silica. In addition the study is extended to the effect of fumed silica on mechanical properties of silica/EPDM nano composites.

2. Experimental

Ethylene propylene Diene Monomer EPDM (type Keltan 6405 , KSA) ,fumed silica (type , XYSIL200 , Henan Xunyu Chemical Co , China) , other ingredients including diocty1phthalate , (DOP), Zinc oxide (ZnO) , Stearic acid , Sulfur (S) ,Mercaptobenzothiazole (MBT) and tetramethy1 thiuram tetrasulfide (TMTD) were of industrial grade products.

The EPDM rubber based composites used in the present study were prepared by using the master-batch technique. In this technique all ingredients were mixed , (except both fumed silica and sulfur) , with the raw rubber material . The obtained batch is divided into equal parts, each part was impregnated with the nano size fumed silica (12 nm) in the range , 5- 25 phr and was added in the last step and hereafter sulfur was added . The silica /EPDM composites with the proper ingredients were prepared on a two-roll mill 170mm-dia,working distance 300mm, speed of slow roll 24 rev/min, and gear ratio 1:4.the curing temperature at 448 °k for 20 minutes in the form of a sheet 15x15x0.2 cm^3 and pressure of 4 M Pa for 10 minutes. Transmission infrared spectra of the fumed silica/EPDM composites were recorded at room temperature using FTIR spectroscopy (ALPHA II, Bruker) at a resolution of 2 cm^{-1} in the range 500–4000 cm^{-1}. The Rheological investigation for the present

fumed silica/EPDM nano composites was carried out using a Rotorless Rheometer , Haida Company , China. The conductivity measurements were carried out on samples of nano composites in the form of discs of 0.2 cm thickness and 1.44 cm diameter where the desired areas were coated with silver paste as contact electrodes . The sample was sandwiched between two brass electrodes and inserted in furnace provided with a copper constantan thermocouple for recording temperature . The dc conductivity was carried out in the range from room temperature up to 453 k where the electric current was recorded using Keithley Autoranging Picoameter , type 485 . The tensile behavior of silica/EPDM nano composites was measured using a custom-built, computer-controlled tensile testing machine on strips of 2.8x3.1x20 mm^3 dimensions which mounted horizontally between grips at strain rate of 3.1×10^{-3} $s^{-1.}$

Table (1): Silica/EPDM compounding ingredients

Ingredients	Phr
EPDM	100
Fumed Silica	5 , 10 ,15 , 20 , 25
DOP[a]	3
ZnO	3
Stearic acid	1
MBTS[b]	1
TMTD[c]	2
S	1

a. DOP : diocty1phthalate plasticizer.
b. phr :parts per handred rubber.
c.. MBTS : Benzothiazoyl Disulfide Accelerator .
d. TMTD : tetramethyl thiuram Disulfide Accelerator.

3. Results and Discussion.

3.1 Rheological investigation:

Rheological parameters, focusing primarily on minimum torque (M_L), maximum torque (M_H), curing time (t_{c90}), and scorch time (t_{s2}), play crucial roles in characterizing the vulcanization process of elastomers. M_L reflects the rigidity and viscosity of the unvulcanized elastomer, serving as an indicator of its processability. On the other hand, M_H, represents the maximum torque attained during curing, directly correlating with the modulus of the compound and thereby measuring its stiffness. The difference between M_H and M_L, known as the delta

torque (ΔM), which is often utilized to estimate the crosslink curing time (t_{c90}) signifies the duration required to achieve 90% of the maximum torque (M_H), while scorch time (t_{s2}) refers to the time taken for the minimum torque to increase by two units, providing insights into the initiation of vulcanization. Also, cure rate index (CRI), is determined as follows [23-25].

$$CRI = 100/(t_{c90} - t_{s2}) \quad \ldots\ldots\ldots \quad 1$$

The cure properties of the three formulations are summarized in Table (2). It is clear that both minimum and maximum torque increase with increasing fumed silica concentration In addition the time at 100% of curing increases with fumed silica concentration to 20 minutes, however , this time is the best for vulcanization of the present nano composites.

Table (2): Extracted values of vulcanization parameters

Silica, concentration phr	M_L Kgf.cm	M_H Kgf.cm	ΔM Kgf.cm	T_{s1} min	T_{s2} min	T_{c10} min	T_{c100} min	T_{c90} min	t at M_H min	CRI S^{-1}
5	3.61	15.24	11.63	0.62	0.78	0.62	8.5	4.3	8.5	3.33
15	6.56	14.56	8.0	0.73	1.0	0.63	9.4	5.8	9.4	0.34
25	9.76	18.51	8.75	0.36	1.12	0.25	20.4	11.78	20.4	0.15

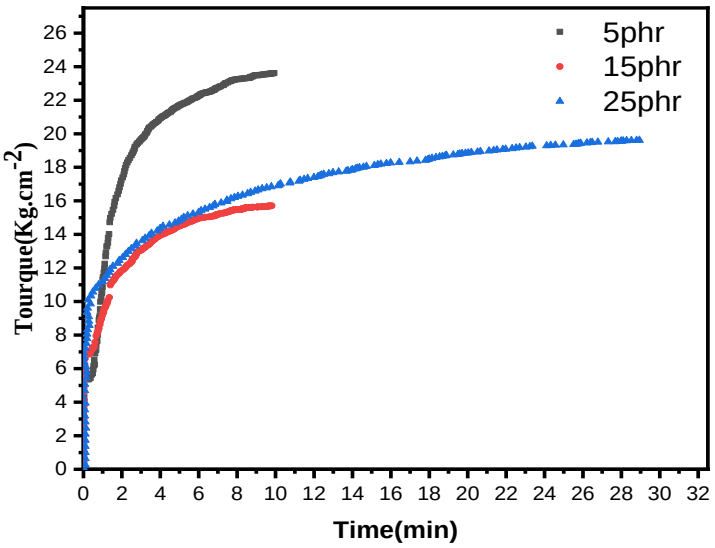

Figure (3.1) Torque versus time for fumed silica/EPDM rubber composites.

3.2 FTIR investigation :

FTIR spectroscopy has been used to analyze the interactions among atoms or ions in silica/EPDM nano composites as shown in Figure 3.2. These interactions may induce changes in the vibrational modes of the polymer electrolyte under investigation.

The absorption peaks of pure EPDM at 2850 cm^{-1} are assigned to the stretching vibration of $–CH_2–$, while at 1640 cm^{-1} is the characteristic absorption of C=C, the peaks at 1460 cm^{-1} is assigned to the in-plane bending vibration of C-H bond, and the peak at 720 cm^{-1} is ascribed to the out-plane bending vibration of C-H bond.

The IR band positions and their assignments are presented in Table (3), which reflects the effect of silica on the chemical structure of the EPDM membrane. FTIR spectra shows no effects of fumed silica on peaks position . As shown from Table (3), the influence of silica loading on the FTIR spectra, including changes in peak intensity and position. Threfore, An inverse relationship between peak intensity and the concentration of silica was observed in the FTIR spectra. This phenomenon suggests a diminishing presence or effect of certain chemical groups or functional moieties within the sample with higher silica concentrations. The reduction in peak intensity indicates alterations in the molecular environment or interactions between silica and other components present in the material. These findings underscore the impact of silica concentration on the spectral characteristics and chemical composition of the composite material.

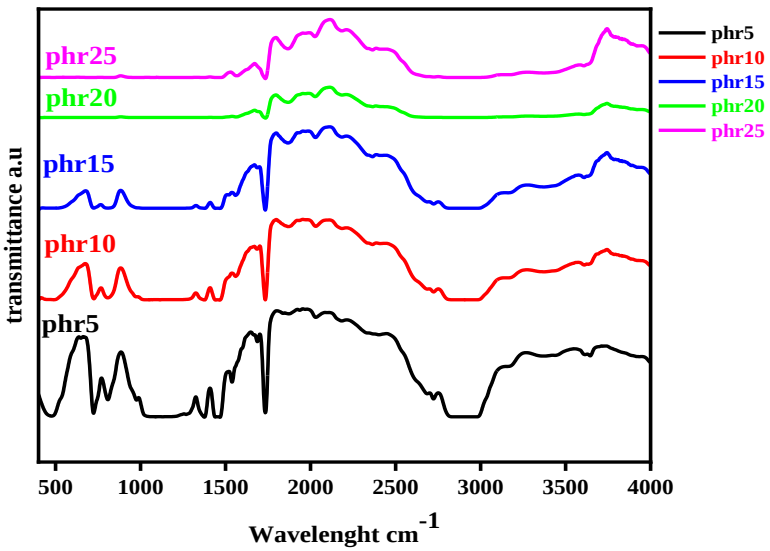

Figure (3.2): FTIR spectra for silica/EPDM nanocomposites with different concentrations of silica

Table (3): FTIR absorption bands position and their assignments for pure EPDM and silica/EPDM nanocomposites

Band assignment	EPDM 0 phr	5phr	10phr	15phr	20phr	25phr
-CH_2-stretching vibration	2850	2850	2850	2850	_	_
C=C absorbtion	1640	1731	1731	1731	1731	1731
C-H inplane bending vibration	1460	1460	1460	1460	_	_
C-H outplane bending vibration	720	720	720	720	_	_

3.3 DC Conductivity investigation:

Figure 3.3 illustrates the temperature dependence of dc conductivity, $\sigma_{dc}(T)$,for fumed silica /EPDM nano composites with different concentrations of fumed silica, 5 - 25 phr.

This behavior of $\boldsymbol{\sigma}_{dc}$ (T) can be described by Mott and Davis equation for conduction in amorphous solids [26] ,

$$\boldsymbol{\sigma}_{dc} = \boldsymbol{\sigma}_o \ \mathbf{exp\ (-\Delta E/KT)} \quad \ldots \quad 2$$

Where ΔE is the thermal activation energy, K is the Boltazman constant, and A is the temperature-independent constant. The activation energy values are extracted using the least squares fitting of equation (2) and listed in table (4) which increases between 0,026and 0.036 eV with increasing fumed silica concentration . The values of room temperature lie conductivity, $\boldsymbol{\sigma}_{RT}$, lie in the range $3.34x10^{-12}$ - $2.9x10^{-11}$ $S.Cm^{-1}$ which refers to unremarkable change of nano composites conductivity with introducing fumed silica filler in such mentioned concentration range (5 -25 phr) and conserving the electrical insulation .

In general, the electrical conduction in fumed silica nano composites can be interpreted as follows. Since electronic conduction in composite matrix occurs through a potential barriers in the polymer chains. Therefore the probability of hopping of charge carriers over potential barriers depends on the barriers heights as well as ambient temperature . Subsequently, the separation distance between silica –silica particle is large enough in the low carbon concentration.

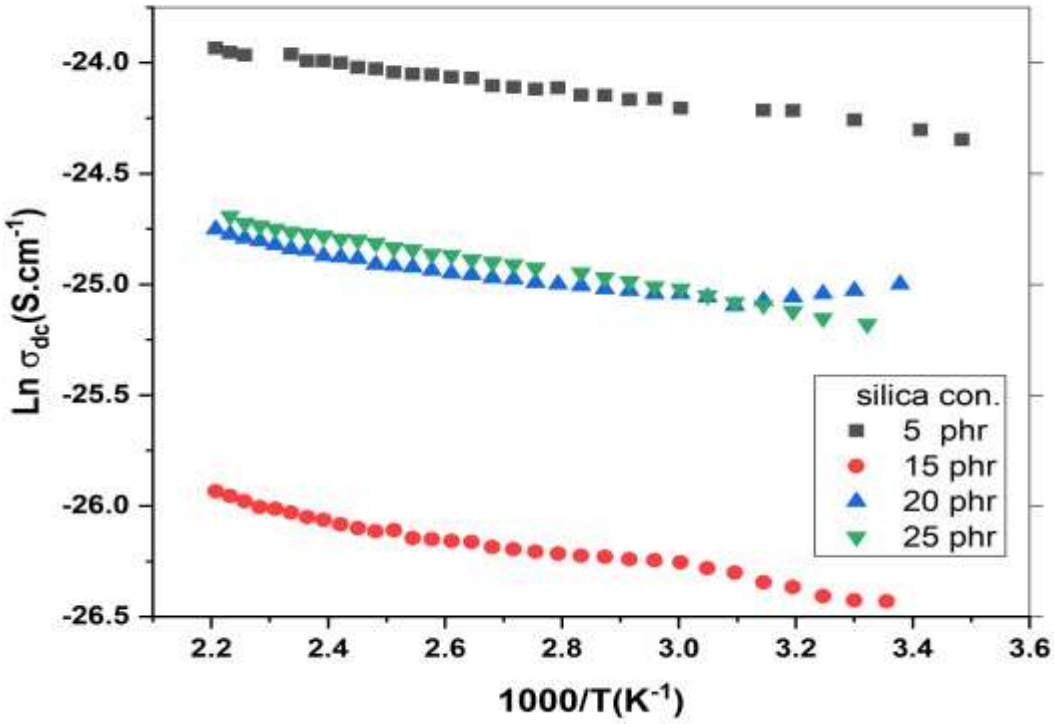

Figure (3.3) Temperature dependence of σ_{dc} for fumed silica/EPDM nanocomposites with different concentrations of fumed silica.

Table 4. Extracted values of activation energies for fumed silica/EPDM nanocomposites.

Fumed silica , (phr)	ΔE , (e.V)	σ_o , ($S.Cm^{-1}$)	σ_{RT}, ($S.Cm^{-1}$)
5	0.026	4.147x10^-11	2.9x10^-11
15	0.033	5.398x10^-12	0.33x10^-11
20	0.030	1.785x10^-11	1.35x10^-11
25	0.036	1.957x10^-11	1.17x10^-11

The nano composite matrix can be regarded as continuous matrix of rubber chains and hence conduction will be controlled by the individual physical properties of EPDM. When the concentration of silica concentration increases, the conduction occurs through silica and EPDM polymer portion connecting silica particles. This means that the conductivity in the conduction

pathways is controlled by the potential barriers heights in polymer chains as well as silica particle – polymer interface.

3.4 Tensile behavior investigation

Figure 3.4 illustrates stress , σ (e) , against strain, e , for fumed silica/EPDM nano composite with different silica concentrations . It's clear that the stress increases linearly with strain up to the yield point, the strain increases with applied stress. The σ (e) behavior clearly shows two regions, the elastic deformation region before the yield point which refers to the composite matrix elastic properties where the region after the yield point illustrates the plastic deformation region

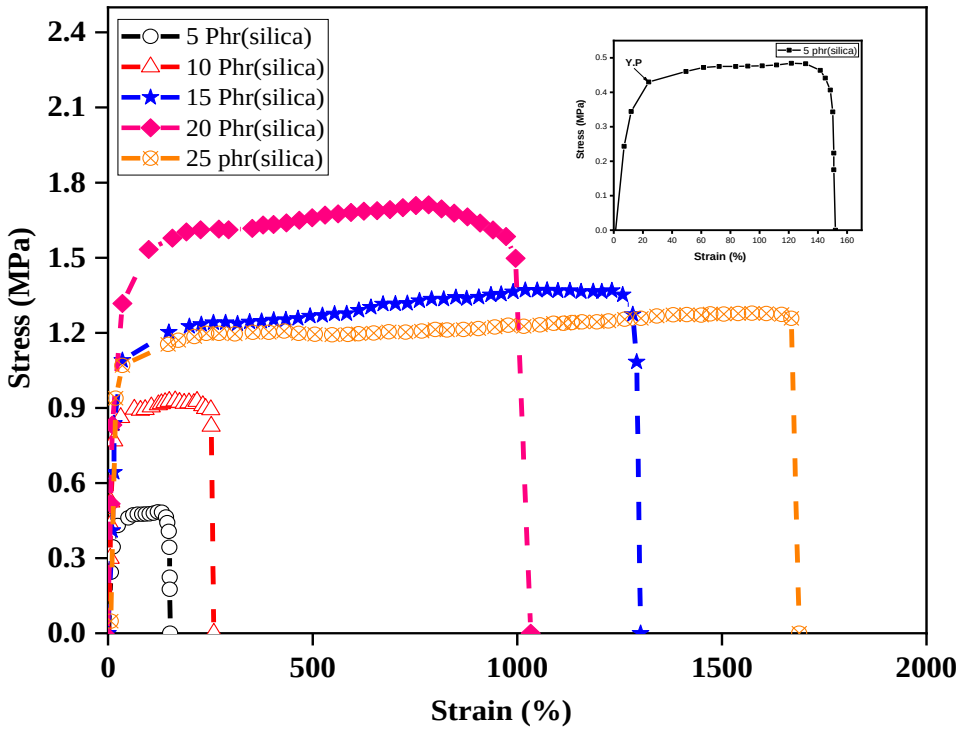

Figure (3.4): Stress – strain relationship for fumed silica/ EPDM nano composites with different concentrations of fumed silica.

The general behavior can be described qualitatively by the following Voigt equation [27] ,

$$\sigma\,(e,\eta) = Ee + \eta\frac{de}{dt} \quad \ldots\ldots\ldots \quad (3)$$

Where , the first term in the right hand in equation 3, represents the nano composite elasticity with elastic modulus , E , the matrix viscoelasticity η and the applied strain rate de/dt. The values of stress σ_e , strain σ_f , and e_f at the fracture point that at yield point σ_y and e_y are obtained for the different concentrations of fumed silica and listed in Table (5) .

Table (5): Extracted tensile parameters σ_y, e_y, E, σ_f, e_f and R for fumed silica/EPDM

fumed silica, phr	σ_{ψ}, M Pa	e_y, %	E, M Pa	σ_{ϕ}, M Pa	e_f, %	**R**, M Pa	$e_f - e_y$
5	0.42	23.9	0.018	0.46	145	6.7	121.1
10	0.86	30.7	0.062	0.89	251	15.06	220.3
15	1.16	198.8	0.007	1.35	1257	104.08	1058.2
20	1.41	157.2	0.013	1.58	972	59.7	814.8
25	1.15	107	0.008	1.25	1669	120.4	1562

Both σ_f and e_f are plotted against fumed silica concentration which clearly shows that σ_f increases to a peak 20 phr of fumed silica concentration and e_f in such fumed silica concentration Figure 3.5 . In addition both σ_y and e_y increase to a peak at 15 and 20 phr respectively with increasing silica concentration Figure 3.5.

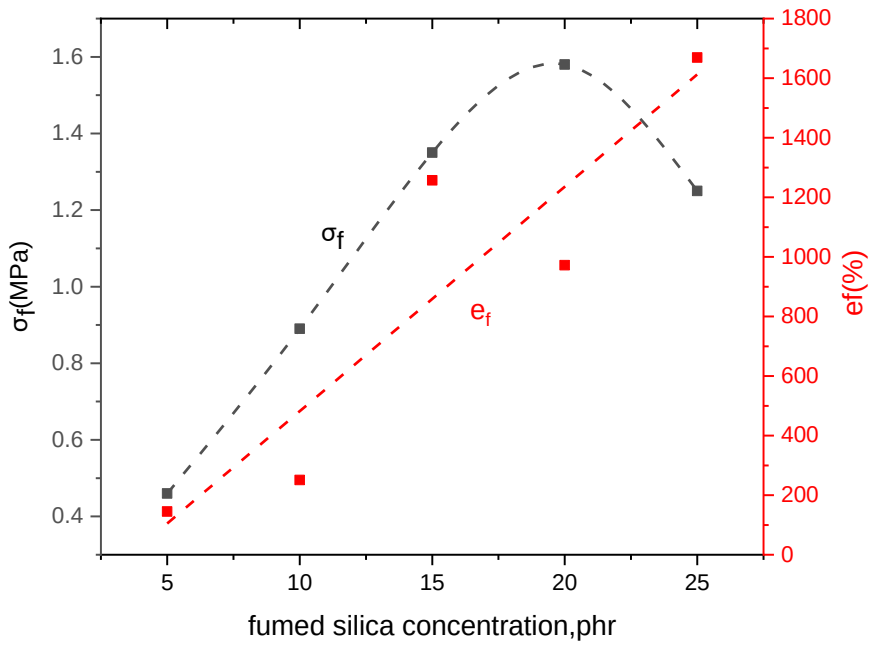

Figure (3.5): σ_f and e_f versus fumed silica concentration.

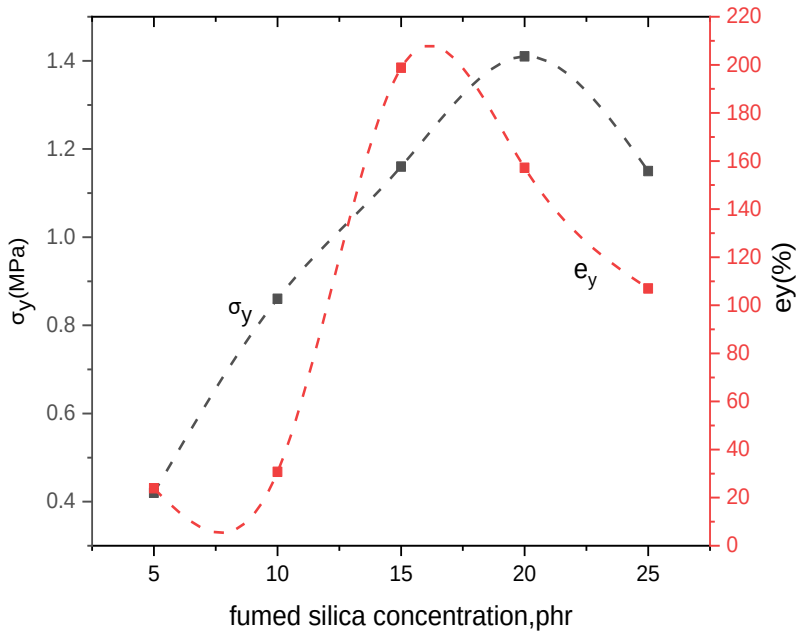

Figure (3.6) : The σ_y and e_y versus fumed silica concentration.

The extracted elastic modulus, E, and the resilience, R, are plotted against fumed silica concentration which illustrates clearly that both E increases to a peak at 18 phr while R increase linearly with increasing fumed silica concentration, Figure 3.7.

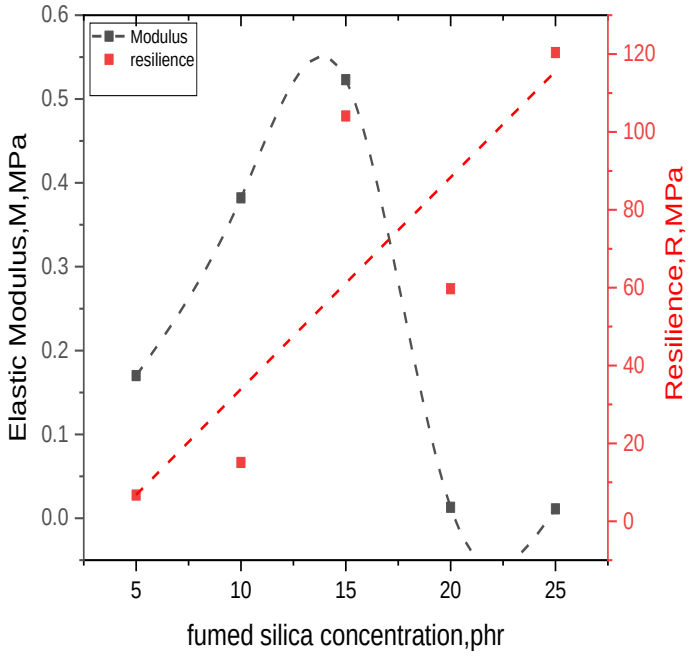

Figure (3.7): Elastic modulus and resilience versus fumed silica.

The general behavior of extension of rubber nano composites can be explained as follows. Since the rubber composite matrix contains large amounts of free volumes created during compounding , this in turns , facilitates the longitudinal strain of chains and reduces the lateral strain until the fracture point. However the incorporation of fumed silica as a reinforcement filler , it will partially occupy the free volume in the matrix . Thus fumed silica reinforcement filler plays an important role in increasing matrix tensile strength and reducing maximum strain.

The increase of fumed silica concentration leads to the increase of both elastic and plastic region to maximum due mainly to the variation of free volume , Table (5) . The presence of polar nano fumed silica (particle size = 12 nm) acts as stress concentrators in relatively high silica concentration (5-25phr) . However , the tensile strength increases , in general , the strain at fracture decrease with increasing fumed silica concentration.

4. Conclusion

The study of fumed silica / EPDM nano composites in the present work illustrates that the conductivity of the nano composites does not show remarkable variation with increasing fumed silica and still lies in the insulation range . In addition the incorporation of fumed silica to EPDM greatly improves the tensile behavior of fumed silica/EPDM nano composites particularly fracture stress and strain as well as resilience .

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