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Study the Effect of adding Natural Rubber and Polymethyl Methacrylate to the Epoxy Resin on the Quantitative Analysis and its Mechanical Properties

***Abstract*-** *From polymer blends can be obtained more useful properties compared with single polymer. In this work, polymer blend (Epoxy (EP): Natural Rubber (NR)) with different ratios of NR (0, 2, 3.5 and 5%wt) and ternary polymer blend with ratio of (Epoxy: 2%NR: 5% wt. PMMA) were prepared. The Mechanical properties were included (tensile, flexural, impact, compression) tests and analytical physical properties (FTIR, SEM) were investigated, and the results show that the elongation values, impact strength and fracture toughness for polymer blend system (Epoxy: NR) were increment with increase natural rubber ratio in the polymer blend system. Whereas fracture strength, young's modulus, flexural strength, flexural modulus and maximum shear stress decreased. The highest values of impact strength and fracture toughness were (0.041KJ/m^2) and (0.321 MPa √m) respectively for polymers blend (95% Epoxy: 5% NR). Ductile fracture in rubber modified epoxy may be produce from the elastomeric nature of rubber which is represents an energy dissipating center.*

Keywords: *Epoxy resin; natural rubber; PMMA; mechanical properties; quantitative analysis*.

1. Introduction

Polymers have become quite important and widely used materials in daily life and industry for the last fifty years. However, for improvement of some properties such as thermal and mechanical stability, polymers industry has developed new production technology depend on blending of a polymer with another component. This kind of strategy allows the limitation of the cost in comparison with the synthesis of new molecules. However, in most of the case, the mechanical properties of the blends are worth than the ones of the pure materials. Controlling the morphology of this blend is the key point of the future development of this type of materials [1]. The properties of polymeric constitutes limit the performance of corresponded blends in addition to the great importance of the morphology of the formed phases that are appear as dispersed spheres, lamellar structures, and co-continuous phases [2] depended on their requirements for a specific application [3,4]. Ratna and Banthia studied the epoxy resins toughened by blending with a suitable liquid rubber, which initially remains miscible with epoxy and then undergoes a phase separation in the course of curing that leads to the formation of a two-phase microstructure [5]. Therefore, the natural rubber is an important class of polymers, it

is a very fundamental material in industry applications due to their unique properties particularly viscoelasticity behavior [6]. Based on the foregoing, it was found that there is an obvious increment in fracture toughness and tensile strength when addition liquid natural rubber (LNR) to epoxy resins [7]. Tan et al. studied the new method to modify the brittleness of the epoxy by adding discrete rubbery phases and then improve the toughness properties. It was found that by adding the rubbery phase into epoxy resin, the toughness of the epoxy was improved. The scanning electron micrograph (SEM) demonstrated the discrete rubbery phases between the epoxy and the rubber particles [8].

In addition from other studies, were found that the tensile strength and Young's modulus for modified epoxy was higher than those of the neat epoxy resin [9,10]. The aim of the work to study some mechanical and analytical physical properties of the epoxy resin through the preparation of binary and ternary polymer blends by adding NR and PMMA to it.

2. Experimental Part

I. Materials

Epoxy resin type (EUXIT 50 KI) was used as the matrix. It was provided from (Al-Rakaez Building

Materials in Amman) made in Egypt Arabic, Epoxy resin of a trade mark (Euxit 50 KI) is a liquid of low viscosity resin and its converted to solid stat by adding hardener (Euxit 50 KII) at ratio of (1:3), which were supplied by Egyptian Swiss chemical industries company. Natural rubber used in the work, RSS-4 Grade Sheets available from North East India also called India rubber or caoutchouc Horizon company (ROMIL-SA select Assured solvent) $(C_6H_5CH_3)$. In this work toluene is used as a solvent for the bulk (NR) which is the pieces of (NR) are immerged in toluene for few days then a solution of (NR) is obtained. Poly (methyl meth acrylate) (PMMA) is powder /self-cure acrylic and (MMA) self-cure liquid use in the research is (Panacryl) type 1-class1-iso 1567 produced in England packaged by Inci Dental in turkey).

II. Preparation of samples

Hand layout technique was used to prepare the polymer blend specimens; a mold of size $(250 \times 250 \times 5)$ mm^3 was made from Aluminum as shown in Figure 1. The inner face of the mold was packed with a layer of thermal nylon papers made of a polyvinyl alcohol substance weighing the epoxy depending on the weight fraction of NR (0, 2, 3.5 and 5%) in polymer blend (Epoxy: NR), and taking into consideration the weight of hardener. The polymer blends were thoroughly mixed by using glass rod to remove any entrap gas bubbles that may be found in this mixture, then the mixture was poured into the mold and allowed to cure for 24h at room temperature (27°C). It is also preparing ternary polymer blend by adding (5%PMMA) and it prepared the same way as previously mentioned. All the specimens were then post cured in an oven at (55°C) for 2h by using electrical oven. Then the sheets cut off and machined according to standard specifications (ASTM) for mechanical tests.

III. Mechanical tests

The samples cut for tensile test according to ASTM D 638 [11]. Microcomputer controlled electronic universal testing machine (model WDW 200 E, China) was used for the testing.

The velocity of (5 mm/min) at ambient temperature was used as conditions for testing. All tests were performed on three samples for the test and the results represent the average data for three samples tested. While flexural strength was tested using a three point method, by using instrument (model WDW 200 E, from China) according to ASTM (D-790-78) at room temperature with velocity (5 mm/min) [12]. The values of flexural strength,

flexural modulus and maximum shear stress, it was calculated by following equations respectively [11]:

$$\sigma_{bend} = \frac{3\,FL}{2wh^2} \qquad (1)$$

$$E_f = \frac{FL^3}{4\,\partial wh^3} = \frac{mL^3}{4wh^2} \qquad (2)$$

$$\tau_{max} = \frac{3F}{4wh} \qquad (3)$$

Where

σ_{bend}: Flexural strength (MPa).

E_f: Flexural modulus (GPa).

∂: The deflection of the beam when a force (F) is applied to it.

τ_{max}: Maximum shear stress (MPa)

F: is the fracture load (N).

L: the distance between the supported points (m).

W: the width of the specimen (m).

H: the thickness of the specimen (m).

According to ISO 179, Impact test was achieved at room temperature [13] using Izod charpy tension impact (Machines Inc, Amityville-New York). Impact strength (G_c) was calculated from the following relationship [11].

$$G_c = \frac{U_c}{A} \qquad (4)$$

Where

G_c: impact strength of the test material (KJ/m^2).

U_c: required energy to fracture the sample (KJ).

A: cross sectional area of the test sample (m^2).

The fracture toughness was calculated from the following relationship:

$$K_c = \sqrt{G_c\,E_f} \qquad (5)$$

Where

K_c: fracture toughness of the test sample (Pa$\sqrt{m}$).

G_c: impact strength of the test material (KJ/m^2).

E_f: Flexible modulus (GPa).

Finally, compression test was done according to ASTM-D 695 [11] at velocity of (5 mm/min) and ambient temperature depending on the thickness of specimens.
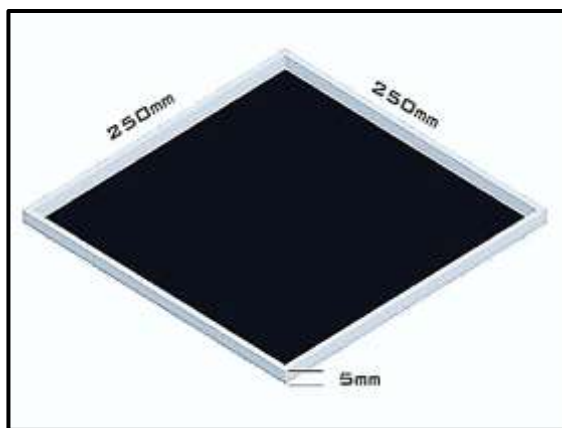
Figure 1: The mold used in the preparation of samples

IV. Analytical physical properties

Fourier Transform Infrared Spectra (FTIR) test
The (FTIR) test is performed according to (ASTM E1252) [14]. By using Fourier transform infrared spectrometer, model (TENSOR 27) made in Germany, by (Bruker Optics Company). It is equipped with a room temperature DTGS detector, mid-IR source (4000 to 400) cm^{-1} and a KBr beam splitter.

Morphology test: To examine surface and fracture surface morphology of polymer material, Scanning Electron Microscope (SEM), model (Tescan VEGA-SB) was used. To achieve good clarity for pictures, all samples are first sputtered with gold from the surface along the edge.

3. Results and Discussion

I. FTIR test result

Mid infrared spectroscopy has been widely used for characterization of organic compounds. This tests are used for fully characterization of epoxy resin and polymeric blends (Epoxy: natural rubber) as a function of natural rubber content in the blend. As well as, the ternary polymer blend (Epoxy: 2%NR: 5%PMMA).

The FTIR spectrum of neat epoxy in the frequency range (400-4000 cm^{-1}) is shown in Figure 2, this spectrum is quiet similar to the reported [14-1٥]. The Characterization absorptions of epoxies involve three locations of the oxirane ring (epoxy groups) bands. The first one is to the C-H tension of the methylene group of the epoxy ring, and is located at 3056.77 cm^{-1} wave number approximately. The second band is attributed to the C-O deformation of the oxirane group, and is located at 915 cm^{-1} wave number, The third band is located at 1181.42 cm^{-1} wave number, and is attributed to C–O stretching of aromatic ring [1٦-1٧].

The IR spectra of epoxy resin reveal the presence of characteristic absorption bands for –C=C–H

aromatic stretching at 3034.80 cm^{-1} wave number The peaks set in range (2850-3000 cm^{-1}) of wave number, which attributed to C-H stretch bond. The peak at higher wave number is attributed to the asymmetric stretch bond of C-H (–CH$_2$ and–CH$_3$) are obtained at (2923.26cm^{-1}) and (2954.84cm^{-1}) wave number respectively, whereas the lower peak is attributed to the symmetric stretch bond of C-H at (2868.52cm^{-1}). The peaks at (1604.99 and 1579.61cm-1) represented to C=C stretching aromatic ring bond. The peak at (1508.45cm^{-1}) corresponds to C-C stretching bond of aromatic ring [14, 18]. The peaks at (1457.45-1362.30cm^{-1}) wave number, was attributed to C-H aliphatic bending bond, as well as, the peaks at (1296.09-1181cm^{-1}) wave number is attributed to C-O stretching of aromatic ring bond, C–O–C stretching of ether group at 1132 cm^{-1}, aromatic stretching at 1106.55cm^{-1}, and the stretch bond of O-C-C was located at (1085.18cm^{-1}) and C-O-C stretch bond at (1036.62cm^{-1}). C–O (epoxy CH$_2$–(O–CH–) oxirane ring stretching vibration at 915cm^{-1} and aromatic absorbance at 826.74cm^{-1} and peak at 758.72cm^{-1} represented to rocking CH$_2$ [14-1٥].

The FTIR spectra of polymers blend (Epoxy-NR) with different ratios of NR (2% and 5%) are shown in Figure 3, from the infrared spectrum of the polymer blend (Epoxy: NR) no other new peak or peak shifts are observed, there is a clear decrease in Peak intensity for all the characteristic peak with addition NR to the polymer blend. As well as when addition of 5%PMMA to polymer blend (Epoxy+2%NR), it is clear from infrared spectrum (Figure 4) of the polymer blend, do not reveal any new peak or peak shifts, there is only decrease in peak intensity for all the characteristic peak, and that may be related to the increase in degree of cross link of epoxy polymer blend [14 and 1٧].
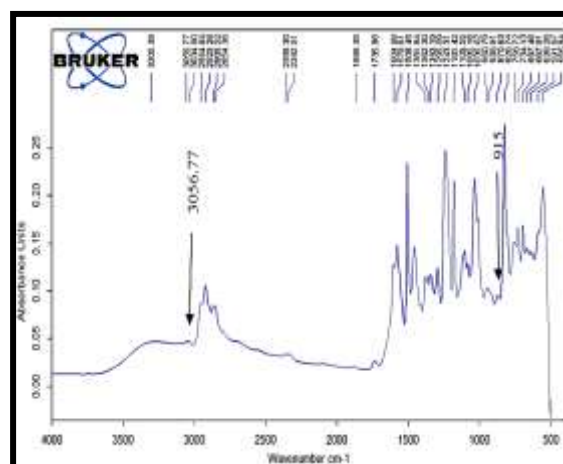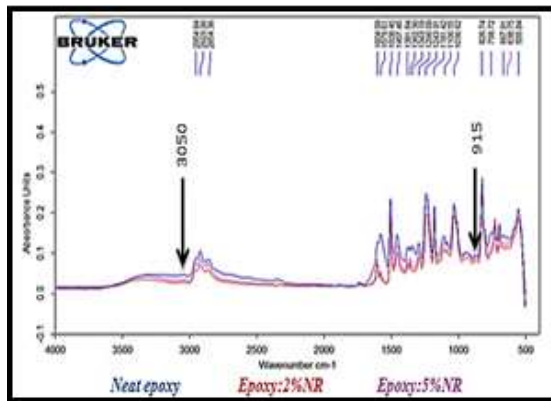

Figure 2: FTIR spectrum for neat epoxy

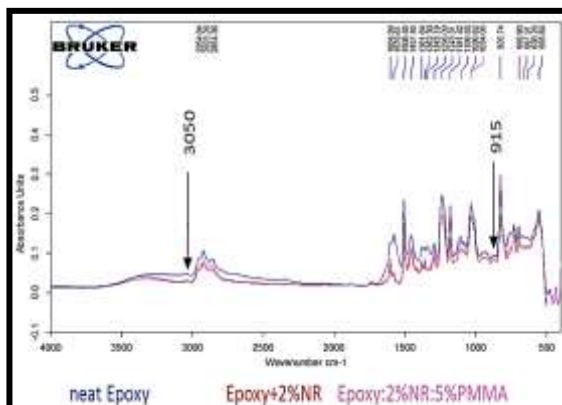**Figure 3: FTIR spectrum of (Epoxy-NR) polymer blend as a function of NR**

Figure (4) FTIR spectrum of neat epoxy, Epoxy+2%NR and Epoxy:2%NR:5%PMMA Polymer blend

II. *Mechanical test* result

Figure 5 shows the (stress–strain) curves of the polymeric blends (Epoxy: NR) with different ratios of NR (0, 2, 3.5 and 5%wt). It has been observed from these curves there are similar behaviors in the (stress-strain) curves when addition of NR to epoxy resin, except it was found that the behavior change from hard and strong for pure epoxy resin to strong and tough with increasing percentage ratios of NR in polymer blend content. Furthermore Figure 5 shows that pure epoxy resin endures a maximum stress at fracture (49MPa). In addition, lower value of % elongation at break (9.21%) as compared to polymeric blends samples. The polymeric blend (95% Epoxy: 5% NR) could bear minimum stress (21 MPa) with maximum elongation (10.11%), this result suggests that the ductile fracture in rubber modified epoxy may be produce from the elastomeric nature of rubber which is represents an energy dissipating center[6 and 7].
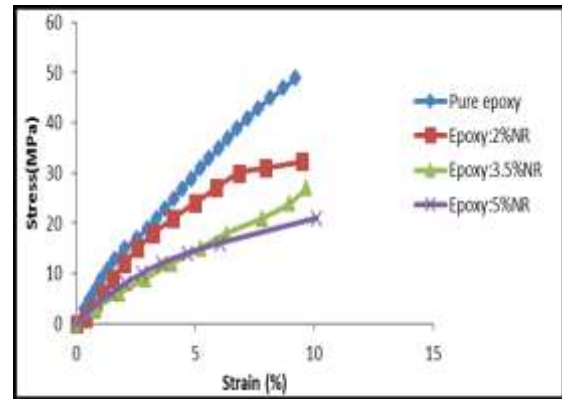

Figure 5: Stress-strain curve for polymeric blends (Epoxy–NR) as a function of natural rubber content in the blends

Figures (6, 7 and 8) shown that, fracture strength values, young modulus and percentage elongation at break respectively for polymeric blends (Epoxy: NR). It was noticed from these figures that the fracture strength and young's modulus for polymeric blends   (Epoxy: NR) decrease with increase NR content in blend and reach to minimum values (21 MPa) and (0.60GPa) respectively at (5%) ratios of NR. While the elongation at break increased with addition of NR and become nearly stable with increase of NR in the blend.
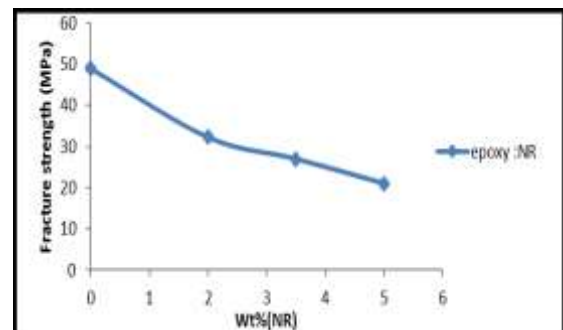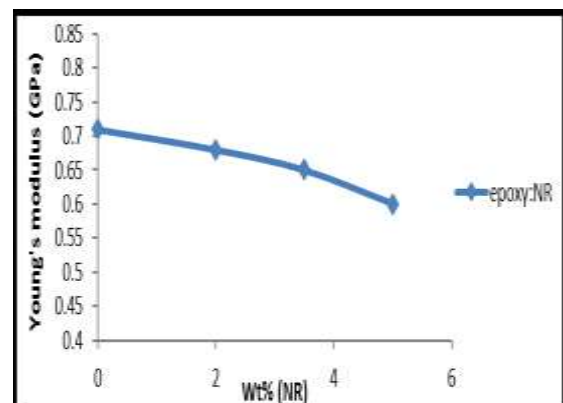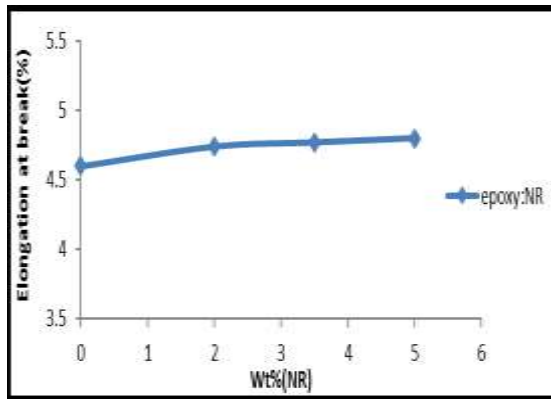

Figure 6: Fracture strength values for binary polymeric blends (Epoxy–NR) as a function of content ratio of natural rubber in the blends

Figure 7: Young's modulus for binary polymeric blends (Epoxy –NR) as a function of content ratio of natural rubber in the blends


Figure 8: Elongation percentage at break for binary polymeric blends (Epoxy –NR) as a function of content ratio of natural rubber in the blends

Figures (9, 10 and11) show the comparison between the samples of the best polymer blend (Epoxy: 2%NR) and ternary polymer blend (Epoxy: 2%NR: 5%PMMA) in terms of the Fracture strength values, Young's modulus values and Elongation percentage at break respectively.

The results of these figures it is showed that the addition of (5%PMMA) to the binary polymer blend (98%Epoxy: 2%NR) gives the higher values of each of fracture strength and Young's modulus except the elongation percentage at break which was lower as compared with the binary polymer blend (98%Epoxy:2%NR), and that may be related to the molecular chain of poly methyl methacrylate (PMMA), when substation of the meth group and methacrylate group on every other carbon atom of the main carbon chain of PMMA material, provides considerable satiric hindrance and this makes polymer blend (Epoxy: NR: PMMA) rigid and relatively strong [11 and ١٧].
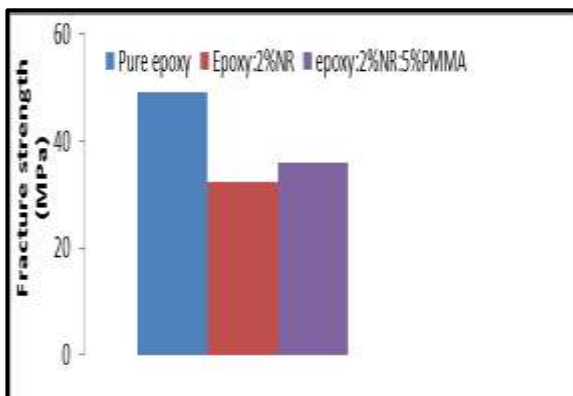
Figure 9: Comparison of fracture strength of pure Epoxy with binary and ternary polymer blends
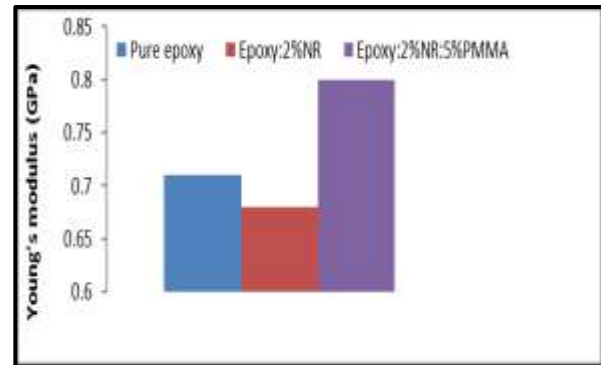

Figure 10: Comparison of young modulus of pure Epoxy with binary and ternary polymer blends
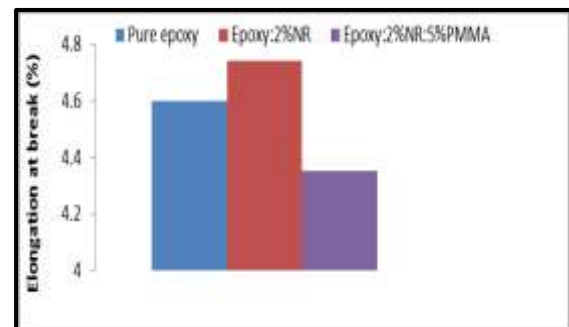
Figure 11: Comparison of Elongation at break of pure Epoxy with binary and ternary polymer blends

III. Flexural test result

Figures (12, 13 and 14) show the effect of addition different ratios of NR (0, 2, 3.5 and 5%wt) on the values of flexural properties (flexural strength, flexural modulus and maximum shear stress) of the polymeric blends (Epoxy: NR) respectively. It has been observed from these figures, there are decreases in values of Flexural properties with increasing percentage ratios of NR in polymer blend content, and this was connected with the natural NR which have low elasticity modulus as compared with Epoxy resin [11].
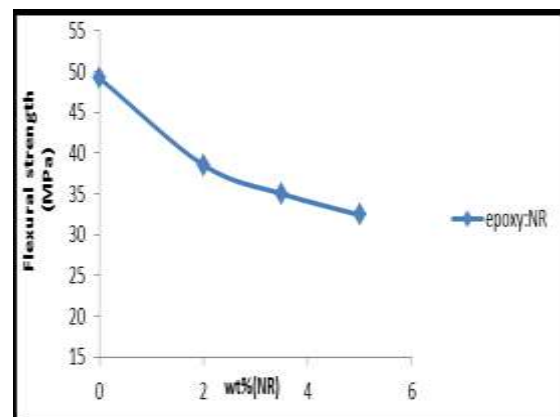
Figure 12: Flexural strength for binary polymeric blends (Epoxy–NR) as a function of content ratio of natural rubber in the blend
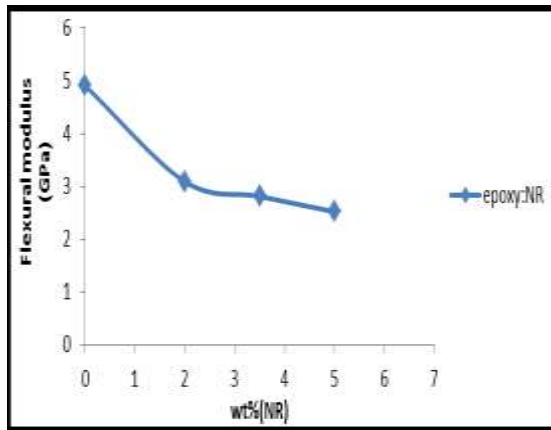
Figure 13: Flexural modulus for binary polymeric blends (Epoxy –NR) as a function of content ratio of natural rubber in the blend
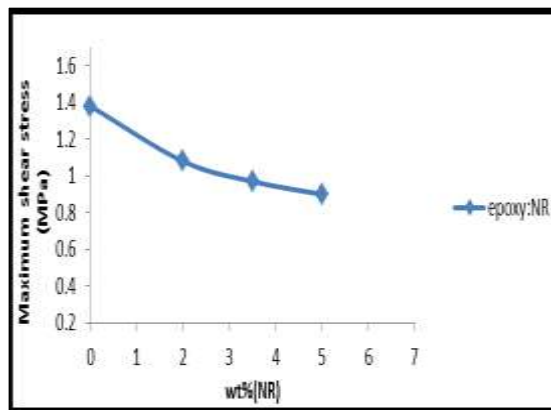

Figure 14: Maximum shear stress for binary polymeric blends (Epoxy –NR) as a function of content ratio of natural rubber in the blend

Figures (15, 16, 17) show the comparison between the samples for the best polymer blend (98%Epoxy: 2%NR) before and after addition 5%PMMA to this polymeric blend, in terms of the Flexural properties respectively. These figures it was show that the addition of (5%PMMA) to the best polymer blend ratio, that it is clear there is decrease in the flexural properties as compared with the epoxy and binary polymeric blend.
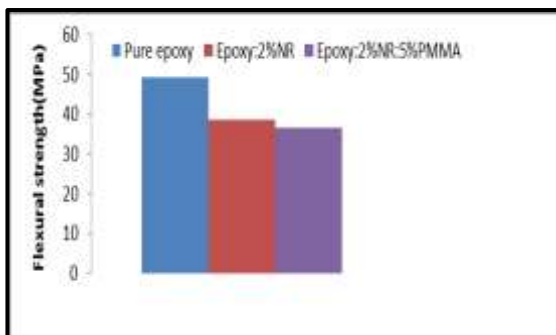

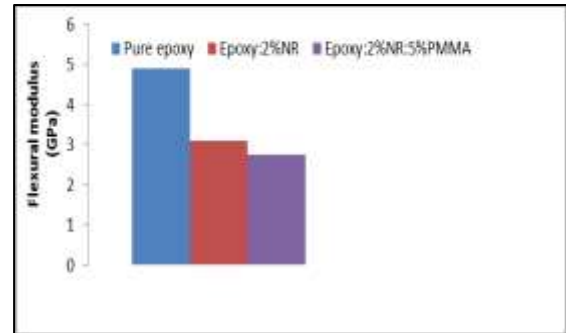**Figure 15: Comparison of Flexural strength of pure Epoxy with binary and ternary polymer blends**

Figure 16: Comparison of Flexural modulus of pure Epoxy with binary and ternary polymer blends
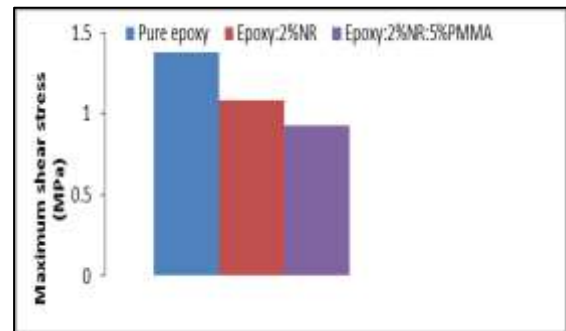

Figure 17: Comparison of Maximum shear stress of pure Epoxy with binary and ternary polymer blends

IV. Impact Test

Figures (18and 19) represents impact strength and fracture toughness curves of the polymer blend (Epoxy: NR). It has been observed from these figures that there is increases in impact strength and fracture toughness with increasing percentage ratios of NR in polymer blend content, so the polymer blend (Epoxy: 5%NR) is the better result of impact strength and fracture toughness than the other ratios in the blend.

Figures (20 and 21) show the comparison between the polymer blend (98%Epoxy: 2%NR) before and after addition 5%PMMA to ternary polymer blend (98%Epoxy: 2%NR: PMMA) in terms of the impact strength and fracture toughness respectively. The results of these figures show that the binary polymer blend (98%Epoxy: 2%NR) have the higher values that related to the nature of rubber phase which behavior as plasticizer material, this can occurs when little amount of rubber is presented in the epoxy resin, or act as flexible material, and this can occurs when natural rubber are incorporated with epoxy resin. Therefore, these two effects lead to increasing toughness of material during the deformation stage, which occurs under shear load, based on the foregoing literatures [19].
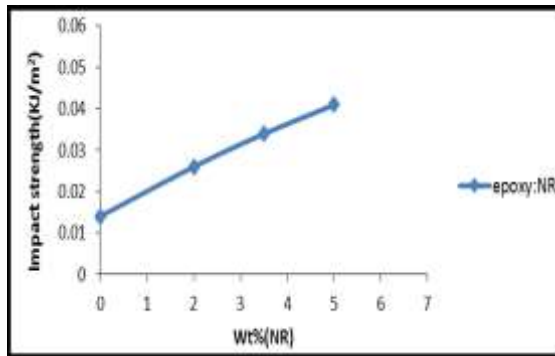
Figure 18: Impact strength for binary polymeric blends (Epoxy –NR) as a function of content ratio of natural rubber in the blend
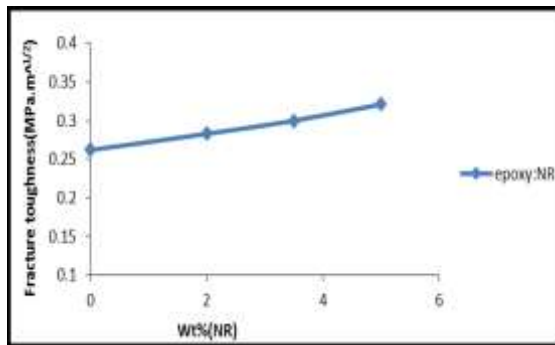

Figure 19: Fracture toughness for binary polymeric blends (Epoxy –NR) as a function of content ratio of natural rubber in the Blend
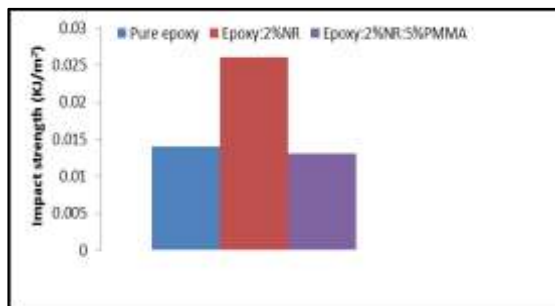

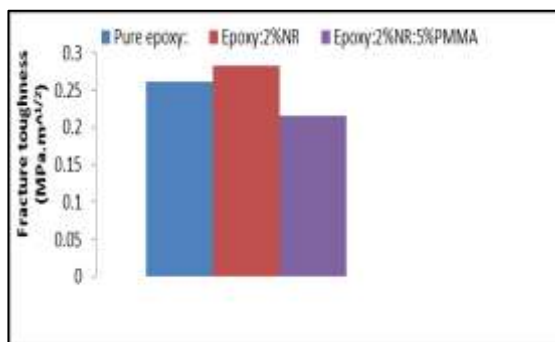**Figure 20: Comparison of Impact strength of pure Epoxy with binary and ternary polymer blends**

Figure 21: Comparison of Fracture toughness of pure Epoxy with binary and ternary polymer blends

V. Compression Test

Figure 22, shows that the compressive strength in pure epoxy is higher than that of other ratios of polymer blend (Epoxy: NR), while Figure 23, shows, that is some improvement in the compressive strength when added 5%PMMA to the polymer blend (Epoxy – 2%NR) and that may be related to the molecular chain of poly methyl methacrylate (PMMA), when substation of the meth group and methacrylate group on every other carbon atom of the main carbon chain of PMMA material, that provides considerable satiric hindrance and this makes polymer blend (Epoxy: NR: PMMA) rigid and relatively strong [11 and 17].
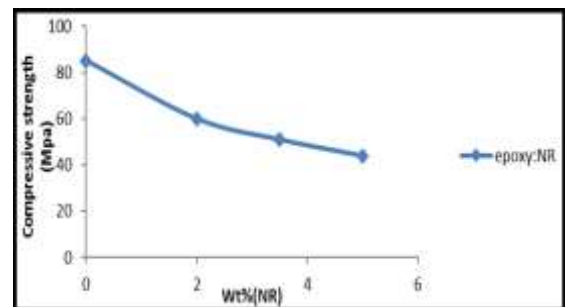

Figure 22: Compressive strength for binary polymeric blends (Epoxy –NR) as a function of content ratio of natural rubber in the blend
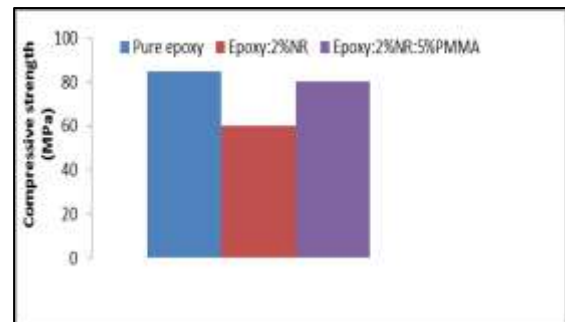

Figure 23: Comparison of Compressive strength of pure Epoxy with binary and ternary polymer blends

VI. Morphology Results

Generally, there are many effects such as component melt viscosities, components ratio and processing conditions are governed the morphology of immiscible polymer blends. The connection between the mechanical properties and morphology for the modified epoxy resins, can be illustrated through micrographs of scanning electron microscope (SEM) of the fractured surfaces of binary polymeric blends (Ep: NR) and ternary polymeric blends (EP: 2%NR: 5%PMMA) at different magnification were recorded in Figure (24). From this figure, it was observed a co-continuous morphology, for both systems of polymeric blend. These results showed

that there are no phase separated domains in the epoxy system and confirmed the formation of homogeneous inter cross-linked network structure's which appear similar to the reference [20] as well as the SEM shown different morphologies with various contents of polymers blend. The fractured surface of binary polymer blend (Epoxy: NR) (Figure 24 a and b) showed homogeneous microstructure micrographs, which show a smoother fractured surface, and this indicate to the better interfacial adhesion between epoxy resin and natural rubber, which appear similar to the reference [21]. Whereas the fracture surface of ternary polymer blend (EP: 2%NR: 5%PMMA) (Figure 24 c and d) exhibited a heterogeneous morphology, a co-continuous morphology of two-phase structure, it was broken down into rough structures, the SEM micrographs show cavities structure, this indicates a weak interfacial adhesion between the components of polymeric blends. [In the end, the morphological analysis of the fractured surface showed rubber dispersions in the sub-micrometer size, leading to improvement in impact toughness, which reached to 123 %]. S. K. Tan et al; reported the role of rubber particles as energy dissipating center in the epoxy structure [8]. Therefore, the morphology of the modified epoxy network by scanning electron microscope can be showed the toughening mechanism, which is attributed to the rubber cavitation, followed by plastic deformation of the matrix resin as, reported in [7 and 8].
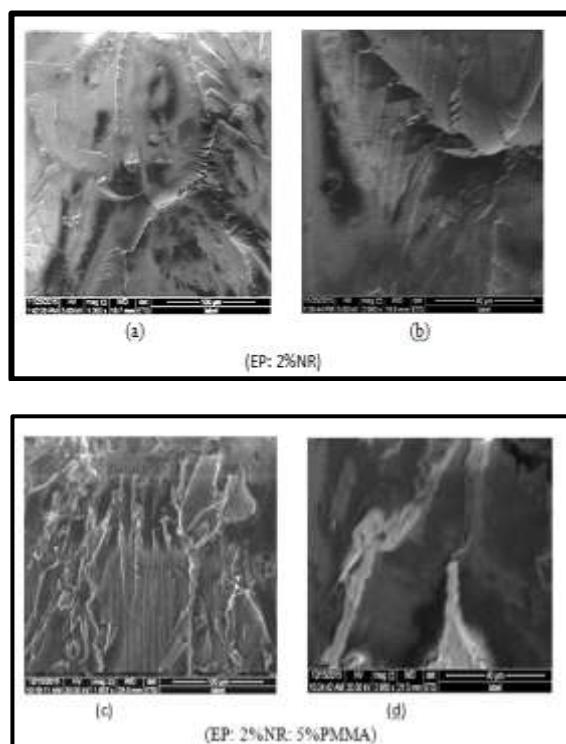
Figure 24: SEM micrographs of the fracture surfaces of (a) (EP: 2%NR) (1000×); (b) (EP: 2%NR) (3000×); (c) (EP: 2%NR: 5%PMMA) (1000×); (d) (EP: 2%NR: 5%PMMA) (3000×)

4. Conclusion

In the present work, attempts are made to study an epoxy resin with desirable properties by adding NR and PMMA to it. It has been observed that addition natural rubber at different weight ratios (2, 3.5 and 5%) to the neat epoxy. This modification has enhanced some mechanical properties of the blend such as impact strength and fracture toughness, improvement in impact toughness reach to 23 %. So it can be concluded that energy dissipating center represented by natural rubber leads to the ductile fracture for the rubber modified epoxy as well as the addition 5% PMMA to polymer blend (epoxy: 2%NR) which gave the best mechanical properties represented by Young's modulus, tensile strength and compressive strength.

Acknowledgement

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