Study of the Properties of Epoxy Nano Composite Reinforced with Different Weight Percentages of Antimony Trioxide

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Abstract

In this study the effect of antimony trioxide nano powders (Sb_2O_3) with weight percentages of (2, 4, 6, 8 and 10 wt%) on some properties of epoxy resin was studied. The nanocomposites are fabricated by hand lay-up method. Some tests are performed such as: hardness, impact strength, thermal conductivity, water absorption, polarization light microscope and thermal stability. It is observed that mechanical, thermal and physical properties of epoxy nanocomposite are modified compared to neat epoxy and found that best weight percentage is (10% Sb_2O_3). The values of hardness, impact strength, thermal conductivity and thermal stability increase ((75.21-79.02), (7.2823-19.0407 KJ/m^2), (0.37957- 0.55438 $W^o.C^{-1}.m^{-1}$) and (17.75- 44% at 500 ℃)), respectively, while the values of weight gain and diffusion coefficient decrease ((0.3533-0.3091 at 10 weeks) and ((2.482985783-1.577478842) $\times 10^{-3}$ m^2/sec)) with the increase of the reinforcement percentage. The hand lay-up method is homogeneous and the surfaces of samples are coherent according to polarization light microscope images.

Keywords: Epoxy Resin, Nano Antimony Trioxide Sb_2O_3, Thermal Properties, Mechanical Properties, Water Absorption, Polarization Light Microscope.

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دراسة خصائص متراكب الإيبوكسي النانوي المدعم بنسب وزنية مختلفة من ثلاثي أوكسيد الأنتيمون

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الخلاصة

تمت في هذا البحث دراسة تأثير أضافة مسحوق ثلاثي أوكسيد الأنتيمون النانوي (Sb_2O_3) وبنسب تدعيم وزنية (2, 4, 6, 8 & 10 wt%) على بعض خصائص راتنج الإيبوكسي. أستخدمت طريقة القولبة اليدوية لتصنيع المتراكبات النانوية. تم تنفيذ بعض الفحوصات مثل: الصلادة و متانة الصدمة و التوصيل الحراري و امتصاصية الماء و المجهر الضوئي المستقطب والاستقرار الحراري. لوحظ أن الخصائص الميكانيكية والحرارية والفيزيائية لمتراكبات الإيبوكسي النانوية حُسنت مقارنة بالايبوكسي غير المدعم ووجد إن افضل نسبة وزنية هي ($10\%\ Sb_2O_3$). قيم الصلادة ومتانة الصدمة والتوصيل الحراري والاستقرار الحراري تزداد ((75.21-79.02) و ($7.2823\text{-}19.0407\ KJ/m^2$) و ($0.37957\text{-}0.55438\ W^{o}.C^{-1}.m^{-}$) و (^{1}) و (17.75- 44% at 500 ºC))، على التوالي بينما قيم الربح بالوزن ومعامل الانتشار تتناقص ((0.3091-0.3533 at 10 weeks) و ($2.482985783\text{-}1.577478842) \times 10^{-3}\ m^2/sec$)) على التوالي مع زيادة نسبة التدعيم. إن عملية القولبة اليدوية كانت متجانسة وسطح العينات متماسك تبعاً الى صور المجهر الضوئي المستقطب.

الكلمات المفتاحية: راتنج الإيبوكسي، ثلاثي أوكسيد الأنتيمون النانوي، الخواص الحرارية، الخواص الميكانيكية، امتصاصية الماء و المجهر الضوئي المستقطب.

Introduction

A composite material can be defined as a mixture consisting of two or more substances of different specifications insoluble in one another to obtain a material whose physical and chemical property differ from the properties of the original materials where the properties of the resulting material are better than their components [1, 2]. The composite material consists of two phases: matrix phase and reinforcing phase, matrix phase may be a metal, ceramic or polymer and reinforcing phase may be particles, fibers, flakes, fillers or laminates [3, 4]. The interest in polymer-based composites has increased because of the specifications of this group of chemical materials, particularly applications using high quality resistance. The most

important materials used as base materials are polymer materials of all three types thermoplastic, thermoset and rubber [5]. The real challenge for most of the materials engineers is modification or development of new composite materials where there is an increase demand for advanced materials with better properties to meet new requirements. One of the approaches to develop new class of polymer is modification of its matrix by addition of powders of different sizes such as antimony trioxide (Sb_2O_3) to achieve the required mechanical properties [6]. Antimony trioxide is an important inorganic compound because of its mechanical and thermal properties where it is used as a flame retardant in engineering materials. It has chemical formula (Sb_2O_3) or (Sb_4O_6) depending on its internal structure, where the cubic structure is colorless, while the orthorhombic structure is white, the cubic antimony trioxide is stable under temperature 570ºC and the orthorhombic is stable above temperature 570ºC [7].

The aim of this study is to manufacture a nanocomposite by adding antimony trioxide to epoxy resins with different weight percentages and to study the effect of these different weight percentages on some mechanical, thermal and physical properties.

Experimental Part

1. Materials

1.1. Matrix Material

Epoxy resin is a low-viscosity transparent liquid that turns into solid state at room temperature after the addition of the cured material with a weight percentage (1: 2). Epoxy resin and hardener were imported from (Sikadur-52 Company, USA).

1.2. Reinforced Material

Antimony trioxide nanopowder is supplied from (Hongwu International Group Ltd, China) with purity 99.5%, particles size (20-30) nm and orthorhombic phase (Valetinite) which has white color.

2. Synthesis

The samples are formed by hand lay-up molding of epoxy resin with weight percentages (2, 4, 6, 8 and 10 wt.%) of Sb_2O_3. The samples are molded according to (ASTM DI-2240) for

hardness test, (ISO-197) for impact strength, (ASTM C20 – 00) for water absorption and polarization light microscope and Lee's disc for thermal conductivity with a tablet shape has a width (40 ± 0.2 mm) and thickness (4.0 ± 0.1 mm). The weight of samples for thermogravimetric analysis (TGA) testing was 17mg.

3. Tests

The hardness was estimated by (Durometer Hardness) type (Shore D) manufactory by (TIME GROUP INC./ ITALY) Company. The impact strength was examined by a (US-made) instrument supplied by (Testing Machines Inc. (TMI)). The thermal conductivity was estimated by Lee's disk test which was supplied by (Griffen and George Company). The surface of samples was imaged by using device type (Leica DM2500 P) supplied by Leica Microsystems company. The thermal degradation was studied using thermogravimetric analysis using (differential scanning calorimetry), model Linseis STA PT1000.

Result and Discussion

Hardness Test

Figure 1 shows the values of hardness of epoxy without and with additive. It can be seen that the hardness increases with increasing weight percentage of nanoparticles because the matrix transfers some of the applied stresses to the particles and therefore increasing material resistance against the plastic deformation, this is in good agreement with [8].

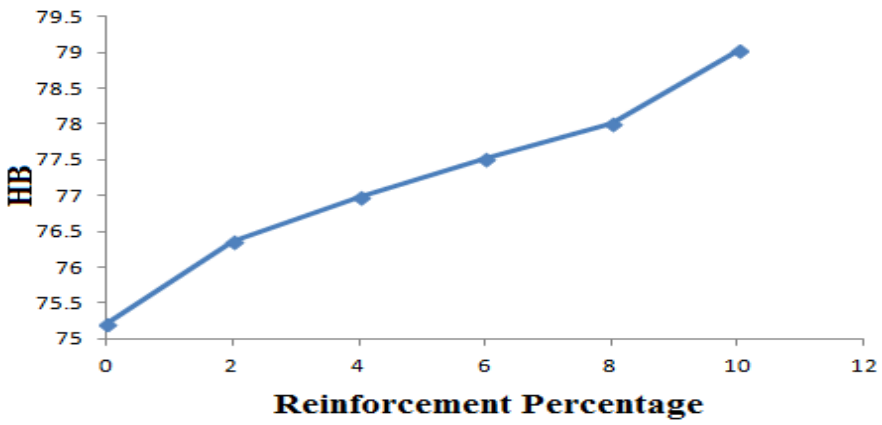

Figure 1: Relationship between hardness and the reinforcement percentage

Impact Strength Test

The energy required to break or rupture samples determine by using Charpy international testing method, this energy and impact strength of material were calculated as follows [9,10]:

$$E = E_1 - E_2 \quad \textbf{(1)}$$

$$I.S = E / bh \quad \textbf{(2)}$$

Where:

E: absorbed energy after impact (Joule).

E_1 and E_2: are initial and final potential energies, respectively.

I.S: impact strength (KJ/m^2).

b: width of the sample (mm).

h: thickness of the sample (mm).

Figure 2 shows the values of absorbed energy in fracture of epoxy without and with additive. In general, the failure occurs in the unreinforced resin material subject to the impact test by breaking the bonds or forces in the polymer because the impact stress caused growing of the initial cracks. In fact, these cracks grow and propagate rapidly towards the interface surfaces separating between the polymer chains because the forces between these chains are represented by van der Waals forces which required low energy to overcome it.

The results show that the increase of the reinforcement percentage leads to increase the impact strength of the samples. This is because the nanoparticles share the matrix phase in the carrying of forces and stresses also prevent the dislocation motion because it penetrates inside the base material and works to fill and reduce the gaps that formed during the molding process, which gave better mechanical properties. This is in good agreement with [11].

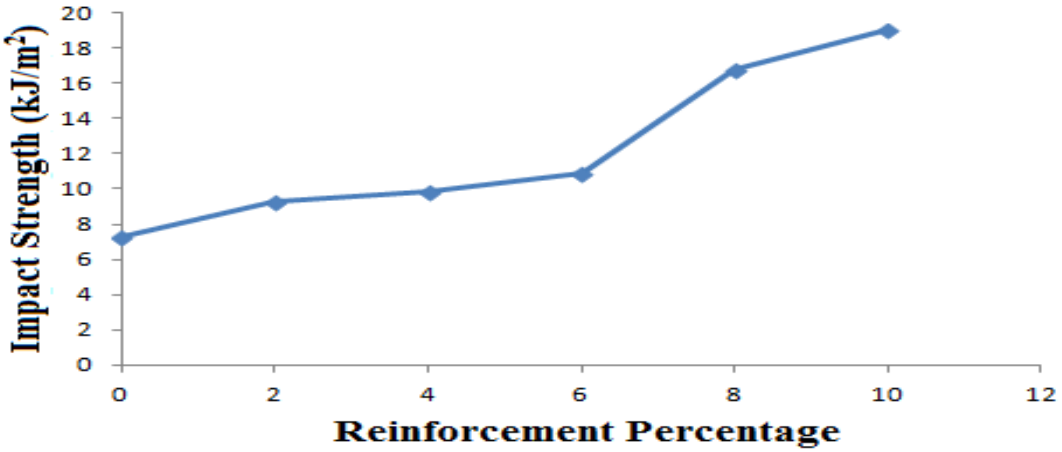

Figure 2: Relationship between impact strength and reinforcement percentage

Thermal Conductivity Test

The Lee's disk consists of three discs (A, B and C) and a heater (H). The sample is placed between the two disks (A and B) and the heater between the two disks (B and C). The device is connected to a power supply and the heat starts transferring from the heater to disc B and then to disk A through the sample. The temperature of the three disks is determined using the thermometers placed inside the disks [12]. The value of thermal conductivity coefficient of the specimen (K) of the sample with thickness (d) and radius (r) is calculated at different potentials across the heater (6 Volt), current which flows through it (0.25 Ampere) and time (1 hour) by using following equation [13]:

$$K\ (T_B-T_A/d_S) = e\ [T_A+2/r\ (d_A+d_S/4)\ T_A+d_S T_B/2r] \quad \textbf{(3)}$$

Where:

d_S, d_A, d_B and d_C: are the thickness of the sample and the disks respectively (mm).

T_A, T_B and T_C: are the temperature of the disks A, B and C (ºC).

e: is the quantity of thermal energy emitted from exposed area of the surface ($W/m^2.ºC$) which is calculated by using following equation:

$$IV= \pi r^2 e\ (T_A+T_B) + 2\pi r e\ [d_A T_A + d_S/2(T_A+T_B) + d_B T_B + d_C T_C] \quad \textbf{(4)}$$

Where:

V: is the different potentials across the heater.

I: is the current which flows through it.

Figure 3 shows the obtained results of measurements of thermal conductivity (W/m.ºC) for epoxy without and with different reinforcement percentage. It has been found that the reinforced with (4, 6, 8 and 10 wt%) nano powders leads to increase the thermal conductivity, because it works to reduce the degree of cross linking between the molecular chains that gives them larger freedom of movement and increase the ability to vibrational motion, which leads to increase the thermal conductivity, this is in good agreement with [14].

The heat passing through the composite material collides with the grains of additives that begin to absorb the heat. This absorption leads to a reduction in the passage of heat through the material, which reduces the thermal conductivity (as occurs with 2 wt%), but after a period of time and at high temperature, these grains begin to vibrate (thermal excitation) due to high temperature causing the rush of heat through the composite material, leading to increase thermal conductivity, this is in good agreement with [15].

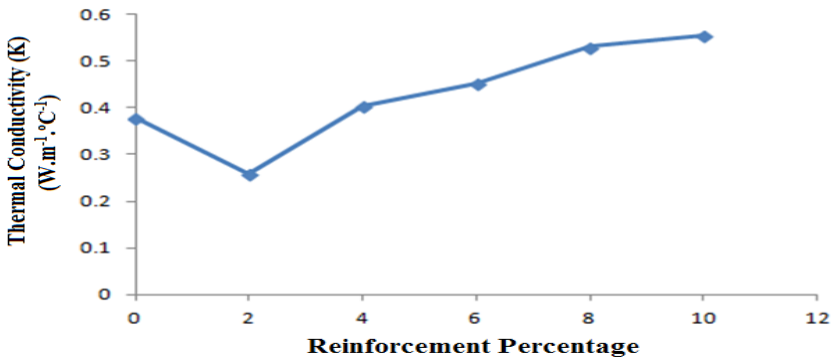

Figure 3: Relationship between K and the reinforcement percentage

Water Absorption Test

The percentage of weight gain for samples prepared after immersion in tap water at room temperature and for periods (10 weeks) was calculated by extracting, drying and weighting

repeatedly every week. The difference in mass before and after immersion is observed using sensitive balance for four decimal digits. Weight gain was calculated using the following equation [16]:

$$\text{Weight gain \%} = ((M_2-M_1)/M_1) \times 100\% \quad \textbf{(5)}$$

Where:

M_1 and M_2: Samples mass after and before immersion (g), respectively.

Also, the diffusion coefficient was calculated by using relationship [17]:

$$Dx = \pi (k_t\, h / 4\, M_\infty)^2 \quad \textbf{(6)}$$

Where:

k_t: Weight gain with time (the slope of the straight line of the curve between weight gain and √time).

M_∞: Maximum weight gain value.

Tables 1 and 2 show the values of the relative percentage of absorption (gain weight) and the diffusion coefficient with the immersion time for 10 weeks for the reinforcement percentage (0, 2, 4, 6, 8 and 10 wt.%), respectively.

Table 1: The relative percentage of absorption of epoxy without and with additives immersed in tap water

Type of additive	Weight gain (%) vs. Time of immersion (week)									
	1	2	3	4	5	6	7	8	9	10
Neat Epoxy	0.1524	0.2155	0.2401	0.2542	0.2724	0.3251	0.3443	0.3527	0.3530	0.3533
Epoxy + 2% Sb_2O_3	0.0809	0.1401	0.1770	0.2140	0.2421	0.2642	0.2902	0.2999	0.3148	0.3173
Epoxy + 4% Sb_2O_3	0.0934	0.1542	0.2024	0.2367	0.2652	0.2849	0.3115	0.3235	0.3268	0.3280
Epoxy + 6% Sb_2O_3	0.0776	0.1248	0.1632	0.1869	0.2196	0.2450	0.2536	0.2786	0.2824	0.2860
Epoxy + 8% Sb_2O_3	0.0827	0.1433	0.1730	0.2122	0.2418	0.2516	0.2800	0.2910	0.2979	0.3030
Epoxy + 10% Sb_2O_3	0.0813	0.1394	0.1840	0.2128	0.2398	0.2563	0.2788	0.2955	0.3024	0.3091

Table 2: The value of diffusion coefficient (Dx) of epoxy without and with additives immersed in tap water

Type of additive	$D_x\ 10^{-13}$ (m^2/sec)
Neat Epoxy	2.482985783
Epoxy + 2% Sb_2O_3	2.28102016
Epoxy + 4% Sb_2O_3	1.961218962
Epoxy + 6% Sb_2O_3	2.182922207
Epoxy + 8% Sb2O3	2.016304746
Epoxy + 10% Sb_2O_3	1.577478842

From table 1, we observe when immersed in water, the weight of the samples increases with increasing immersion time, because the low molecular weight of the water helps it to quickly penetrate through the polymer and fill the gaps, this is in good agreement with [12].

From table 2, we observe a decrease in the values of diffusion coefficient for weight percentage (2, 4 and 10 wt%) because of the reinforcement works to fill the gaps and voids in the sample therefore the difficulty of penetration of water through it, while we observe a increase in the values of diffusion coefficient for weight percentage (6 and 8 wt%) due to defects formed during the molding process these defects helped to penetrate the water is rapid through polymer chains which lead to increase the chemical bond between them. When the density of bonding increases the susceptibility to absorption of polymer decrease, the difference in concentration between the liquid and the area in which will penetrate during increase. The speed of diffusion is higher according to Fick's law of diffusion, this is in a good agreement with [18].

Polarization Light Microscope Test

The topography of the material was studied by using polarization microscope technique. Samples (neat epoxy) and (Epoxy/10% Sb_2O_3) were imaged before and after immersion in tap water for (10 weeks). The microscope images showed that the surface of the material before immersion seems coherent and this is evidence that the process of mixing is homogenous while the microscope images after immersion emergence of some of the voids on the surface the samples in different degrees, this is a proof of the penetration of water into samples as shown in figures (4 - 7).

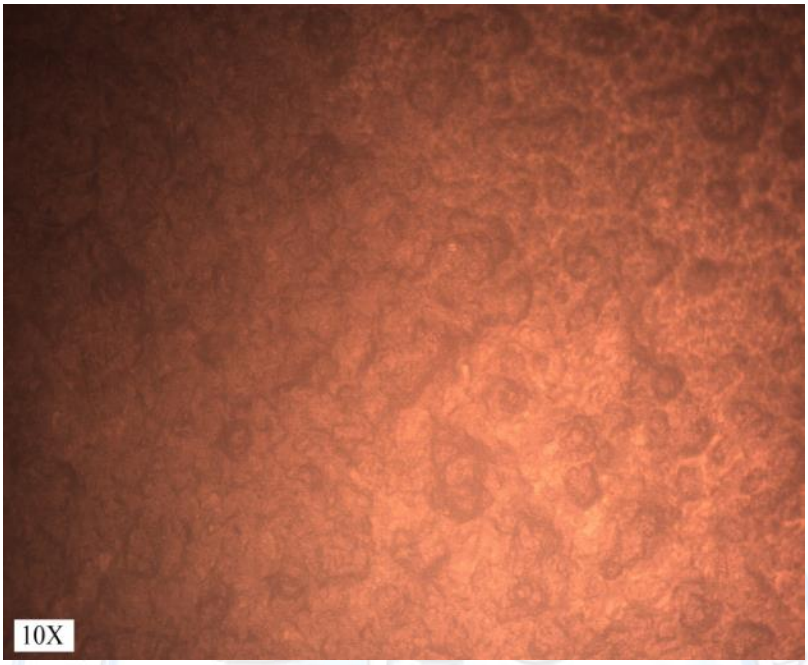

Figure 4: Neat epoxy before immersion

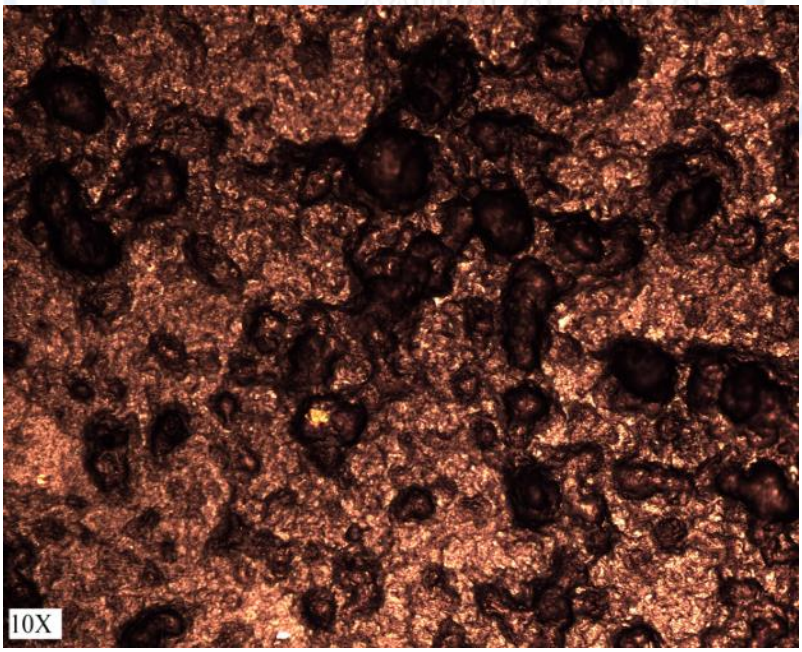

Figure 5: Neat epoxy after immersion

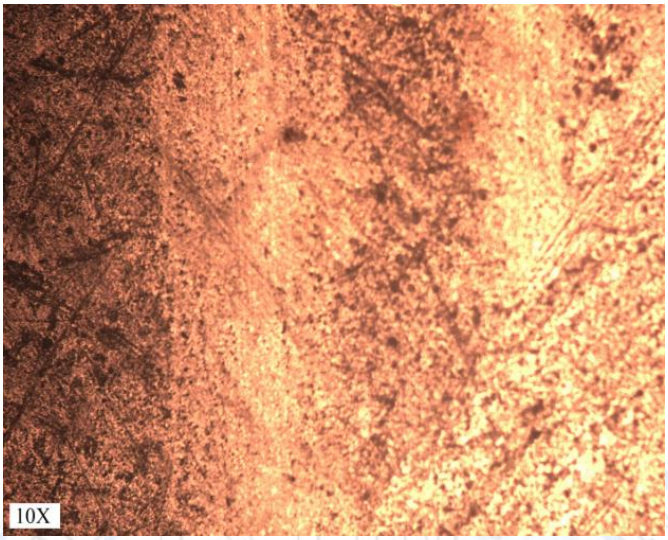

Figure 6: (Epoxy + 10wt.% Sb_2O_3) before immersion

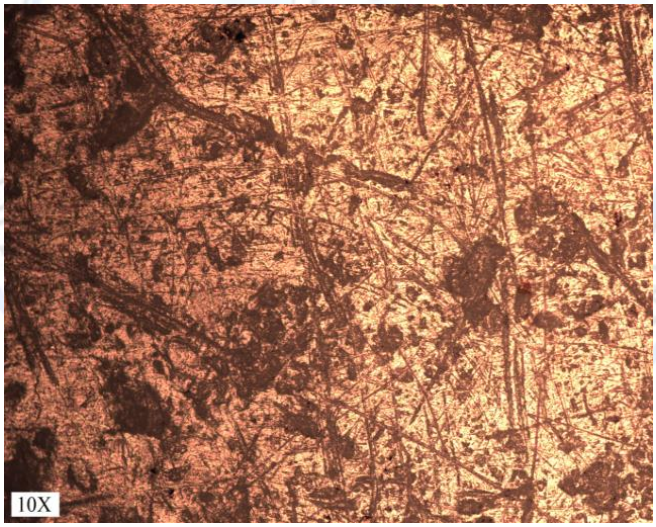

Figure 7: (Epoxy + 10wt.% Sb_2O_3) after immersion

Thermogravimetric Analysis Test (TGA)

TGA is used to know the thermal stability of epoxy resin and the effect of additive on thermal stability for it. Table 3 illustrates the TGA results of epoxy without additive and with (10 wt.%) additive. In both samples a change in peak was observed due to different melting behaviors of composite materials. There is no change in the start of melting thermogram and there was a

marginal increase in its end, this difference is due to that the nanoparticle penetrates inside the base material and works to fill and reduce the gaps that are formed during the molding process, which gave better thermal properties. The weight loss is due to the epoxy resin loses moisture by evaporation process and begins to decompose.

Table 3: Thermogravimetric analysis results of epoxy without and with additives

T ºC	Rel. mass change (%)	
	Neat Epoxy	Epoxy+10% Sb_2O_3
100	100	100
150	100	100
200	92	100
250	81	99
300	77	97
350	60	84
400	34	58
450	27	51
500	17.75	44

Conclusions

Neat epoxy has lower mechanical and thermal properties than (reinforced epoxy) composites. Increased reinforcement percentage help increasing the absorbed energy required to fracture the sample and therefore increases impact strength. Hardness and thermal properties (thermal conductivity and thermal stability) are increased with the increase of weight fracture of epoxy resin for all reinforcement percentage. Weight gain and diffusion coefficient values is decrease with the increase of reinforcement percentage.

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