

## Study of Thermal Behavior for Epoxy and Some of Its Composites in Different Temperatures Using Water Bath Technique

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### Abstract

The aim of this research is to study the property of thermal conductivity for epoxy and some of its composites at room temperature, and various temperatures for various period of immersion using water bath technique. Three samples are prepared by hand lay-up method. The first sample is epoxy resin, the second sample is epoxy resin reinforced with Aluminum powder, and the third sample is epoxy resin reinforced with Al fibers. The volume fraction is 15%. Thermal conductivity is measured for three samples at room temperature (30 °C) using Lee's disc device. Thermal conductivity is measured for samples after immersion in water bath for (30, 60) minutes at (40, 50) °C. Activation energy is measured for samples. Results show generally that reinforcing with Al powder and fibers increases thermal conductivity. Also thermal conductivity increase for samples at 50 °C after immersion in water bath at this temperature as compared with other temperatures. It is found that thermal conductivity, generally, decreases by increasing the period of immersion in water bath whereas thermal conductivity recorded lower values after immersion for 60 minutes. Results shows that sample of epoxy reinforced with Al powder have the highest value of activation energy as compared with other samples.

### دراسة السلوك الحراري للأيبوكسي وبعض متراكباته بدرجات حرارة مختلفة

### باستخدام تقنية الحمام المائي

### الخلاصة

يهدف البحث الحالي الى دراسة خاصية التوصيل الحراري للأيبوكسي وبعض متراكباته بدرجات حرارة الغرفة، وبدرجات حرارة مختلفة ولفترات زمنية مختلفة من الغمر باستخدام تقنية الحمام المائي. تم تصنيع ثلاث نماذج بطريقة القولية اليدوية. تم تحضير راتنج الأيبوكسي وهو النموذج الأول، وتم تدعيم الراتنج بمسحوق الألمنيوم وهو مايمثل النموذج الثاني، وتم تدعيم الراتنج باللياف الألمنيوم وهو مايمثل النموذج الثالث. الكسر الحجمي المستخدم 15%. تم قياس التوصيلية الحرارية للنماذج الثلاثة بدرجات حرارة المختبر (30) درجة مئوية باستخدام جهاز قرص لي لقياس التوصيل الحراري. وتم قياس التوصيلية الحرارية للنماذج بعد غمرها في الحمام المائي لفترات زمنية (30، 60) دقيقة وبدرجات حرارة (40، 50) درجة مئوية. كما تم حساب طاقة التنشيط للنماذج. أظهرت النتائج أن التدعيم عموماً بمسحوق وألياف الألمنيوم أدى إلى زيادة التوصيلية الحرارية. كما زادت التوصيلية الحرارية للنماذج عند درجة حرارة 50 درجة مئوية بعد غمر النماذج بالحمام المائي عند هذه الدرجة بالمقارنة مع الدرجات الحرارية الأخرى. كما وجد أن التوصيلية الحرارية للنماذج تقل عموماً بزيادة الفترة الزمنية للغمر بالحمام المائي حيث سجلت التوصيلية الحرارية قيماً أقل عند الغمر

بالحمام المائي لفترة 60 دقيقة بالمقارنة مع  
قيم التوصيلية الحرارية عند الغمر لفترة 30 دقيقة. وأظهرت النتائج أن نموذج الأيوكسي المدعم  
بمسحوق الألمنيوم امتلك أعلى قيمة لطاقة التنشيط مقارنة مع النماذج الأخرى.

## 1. Introduction

Polymer composites filled with metal are of interest for many fields of engineering. This interest arises from the fact that the electrical characteristics of such composites are close to the properties of metals, whereas the mechanical properties and processing methods are typical for plastics. The achievement of metallic properties in such composites depends on many factors, and it is just the possibility of controlling the electrical and physical characteristics which determines the variety of ranges of their application. The filling of a polymer (Also, two types of polymers thermoplasticpoly (vinyl chloride) and thermosetting epoxy resin) were used as matrices) with metallic particles (Copper and nickel powders having different particle shapes were used as fillers). Results show an increase of both electrical and thermal conductivity of the composites obtained. <sup>(1)</sup>

The addition of conductive filler in a polymer matrix is an effective way to increase the thermal conductivity of the plastic materials, as required by several industrial applications. All quantitative models for the thermal conductivity of heterogeneous media fail for heavily filled composites. The Onductivity

of the filler, its size and shape, and its local amount are, with the degree of previous mixing, the main factors determining the effective conductivity of composites. <sup>(2)</sup>Epoxy resins are Generally used in many electronic packaging applications, but their low thermal conductivity presents a major drawback. Therefore, high intrinsic thermal conductive solid fillers, e.g., metal powders, carbon fiber, graphite, diamond, ceramic particles and whiskers have been incorporated into epoxy resins. <sup>(3)</sup>It is well known that thermal transport increases significantly in the direction of orientation and decreases slightly in the direction perpendicular to the orientation. But most of these studies were confined to the thermal behaviour of neat polymers only and not to their composites. Reports are available in the existing literature on experimental as well as numerical and analytical studies on thermal conductivity of some filled polymer composites. The fillers most frequently used are aluminum particles, copper particles, brass particles, short carbon fiber, carbon particles, graphite, aluminum nitrides and magnetite particles. <sup>(4)</sup>Thermal conductivity is the property that determines the working temperature levels of a material, and it is an important parameter in problems involving steady state heat transfer.

However, it is one of the physical quantities whose measurement is very difficult and it requires high precision in the determination of the parameters involved in its calculation. <sup>(5)</sup>On the other hand, heat transfer is restricted by phonon scattering at the matrix-filler interface. As the loading content of the filler is increased, the polymer filler interfacial area increases, which subsequently increases the phonon-scattering. It was found that, in most cases, phonon scattering reduces the amount of heat transfer in composite material systems <sup>(6)</sup>. Consequently, if the interfacial adhesion is not good and, thus, the extent of phonon scattering is significant in composites, the highly-conductive fillers may decrease the thermal conductivity with increasing filler-loading content. In addition, when the particles have different sizes, the numbers of interfaces along the path through which heat is transferred varies; therefore, composite materials with the same filler content have different values of the thermal conductivity. In short, composite materials have different thermal conductivities depending on the size or loading content of the particles, characteristics of the interfaces, or relative thermal conductivities of the particles and resin <sup>(6)</sup>

**Ramazan and Mamdouh** , were investigated moisture diffusion behavior of aluminum powder-filled epoxy adhesive through utilizing fluid immersion tests under complete immersion in salt solutions with varying NaCl concentrations. Aluminum powder is used in the adhesive for the purpose of improvement of its thermal properties, as demanded in a variety

of industrial applications. They found the aluminum filler content did not affect the moisture diffusivity in

epoxy adhesive significantly but the effect of salt concentration was significant; the higher the salt content in the test solution, the higher the moisture diffusivity in the adhesive <sup>(7)</sup>

**Zhou et al.** were prepared microsized aluminum/epoxy resin composites, and the thermal and dielectric properties of the composites were investigated in terms of composition, aluminum particle sizes, frequency, and temperature. It was found that the aluminum/epoxy composite containing 48 vol % aluminum-particle content possessed a high thermal conductivity and a high dielectric permittivity, but a low loss factor, a low electric conductivity, and a higher breakdown voltage. <sup>(8)</sup>

Application include encapsulation, die (chip) attach, electric cable insulation. In addition aluminum powder is used in the adhesive for the purpose of improvement of its thermal properties, as demanded in a variety of industrial applications <sup>(9)</sup>.

Aluminum has conventionally been used in many composite applications, the industrial waste cement by-pass dust can be considered as non-conventional materials for use as fillers in polymer composites <sup>(10)</sup>The aim of this study is to improve the thermal conductivity of polymers because polymers have a very low thermal conductivity, compared with metals; the choice of filler depends on the end use. Metal fillers will improve

machine ability, hardness and thermal conductivity.

Moreover, is to show the drastic influence of the moisture content on the thermal conductivity and it is investigated the effect of adsorbed water, on the thermal conductivity.

## 2. Experimental part

Hand lay – up method is used to prepare the composite samples. Composite is made using a stainless steel mould having dimensions of (25\*25) cm<sup>2</sup>. Epoxy resin (Quickmast 105: made in Saudi Arabia) is mixed with hardener in ratio of (3:1). The Al powder (that grain size of (90 - 106µm particle fraction size a verge size 100µm) is mixed with resin with volume fraction of (15% V<sub>f</sub>) then the mixture is poured into mould slowly in order to avoid air trapping.

The second composite, seven layers of Al fibers are used. (The Young modulus of Al fiber is 70 GN/m<sup>2</sup> and the density is 2.7 gm/cm<sup>3</sup>) The matrix is poured into mould slowly which is covered the Al fibers. The composites left for (24 hours) at room temperature to get solid samples Then the composites was heated for 50 °C for 3 hour to complete the curing process.

The samples are cut with standard dimensions coincide with (ASTM-D150) for thermal conductivity test. To prevent filler sedimentation and to ensure random spatial distribution within the polymer matrix, silica foam is used for such aim with sample of epoxy reinforced with Al powder. Commercial name is (Aerosid200) the grain size of silica foam is 0.012 µm and the density is 2gm/cm<sup>3</sup> It is used to prevent the precipitation of Al powder and to

get a homogeneous distribution of Al powder in resin. This technique is known as powder dispersion.

## 3. Measurements

Thermal conductivity In this work thermal conductivity is measured. The test is carried out in accordance with Lee's disk. Lee disk manufactured by (Griffen and George company/England), this device consists of three disks of brass with a special heater, as shown in figure {1}. The specimen (S) is placed between two disk (A, B). Disk B shall be contacted to electrical heater (H) and then comes the disk (C) which represents the thermal conductivity device.

The thermal conductivity calculated from the equation:

$$IV = \pi r^2 e (T_A + T_B) + 2\pi r e [d_A T_A + 1/2 d_S (T_A + T_B) + d_B T_B + T_C] \dots\dots (1)$$

$$K(T_B - T_A / d_S) = e [T_A + 2/r \{d_A + (1/4)d_S\} T_A + (1/2r)d_S T_B] \dots\dots (2)$$

Where:

I: (0.25 mA) current value (Ampere).

V: (6 volt) applied voltage (Volt).

r: radius of the disks (mm).

e: heat flow per unit area per second. (W/m<sup>2</sup>. °C).

T<sub>A</sub>, T<sub>B</sub>, T<sub>C</sub> : temperature of the disk A, B, C. (°C).

d<sub>A</sub>, d<sub>B</sub>, d<sub>C</sub> : thickness of the disk A, B, C. (mm).

d<sub>S</sub> : thickness of the sample (mm).

K: thermal conductivity (watt/m.C).

Water bath is used in this research. It manufactured by (Memmert / Germany). In order to get an estimate of thermal conductivity at different temperature (40°C, 50°C) with different time.

Activation energy is calculated by using the following Arrhenius equation:

$$K = K_0 \exp (E_a/RT) \quad \dots\dots (3)$$

Where  $E_a$ ,  $R$ ,  $T$ ,  $K_0$  are Activation energy for samples(KJ/mole), Universal gas constant (8314J/kg.mole.K), Temperature in degree Kelvin and thermal conductivity without

This equation can be reduced into a linear form by taking natural log on both sides.

$$\ln K = \ln K_0 (E_a/R).1/T \quad \dots\dots (4)$$

A plot of  $\ln K_0$  versus  $1/T$  should be nearly linear, and with a slope of  $E_a/R$  using the data in table (1) and determine the activation energy between  $\ln K$  and temperature (40 and 50°C) in 30 and 60 min.

Figure{2} shows samples of this research.

#### 4. Results and Discussion

Thermal conductivity of a composite depends on many parameters including: (1) fiber orientation; (2) moisture content ;( 3) distribution of moisture; (4) void percentage, (5) resin type and (6) filler percentage. The parameters of major influence on thermal conductivity are fiber volume fraction and conductivity properties of both resin and fiber. <sup>(10)</sup>

From table (1) it can be seen that the values of thermal conductivity for both composites are higher than of this for the resin matrix, because Aluminum fibers have a high thermal conductivity than the resin matrix. ( $K = 250 \text{ W/m. } ^\circ\text{C}$ ) for Aluminum.

It is known that the transport of heat in non metal occurs by phonons or lattice vibration. The thermal

resistance is caused by various types of phonons scattering processes, e.g. phonon- phonon scattering, boundary scattering and defect or impurity scattering. In order to maximize the thermal conductivity, these phonon scattering processes must be minimized. Phonons travel at the speed of sound. The scattering of phonons in composites material is mainly due to the interfacial thermal barriers resulting from acoustic mismatch and flaws associated with the filler- matrix interface <sup>(9)</sup>

The thermal conductivity of Aluminum powder/epoxy composites was higher than that of Aluminum fibers/epoxy composites .This can be discussed by the heat transfer from particle to particle is easier because of thinner polymer layers between the particles which increase the conductivity of composite <sup>(1)</sup>. Moreover a good interfacial between Al powder and Epoxy resin which increase the thermal conductivity.

During composite fabrication by hand-layup technique the formation of holes cannot be neglected and may even formation of a thin interfacial gap between the fiber and resin due to inadequate interfacial compatibility acting like air void and in turn reducing the thermal conductivity<sup>(10)</sup>

After immersion in water bath with 40°C for 30 and 60 min. thermal conductivity of all samples decreased in comparison with results of thermal conductivity at 50°C for 30 and 60min,as shown in figure(2). .But the samples that had been already exposed to high temperatures of water for a long

period of time have low thermal conductivity, as shown in table {1}. These results explain the values of thermal conductivity measured at 50° C higher than those measured at room temperature .As the temperature increases the amount of adsorbed water decreases. The decrease of the amount of adsorbed water leads to increase in the thermal conductivity.

The long time of immersion might the water molecules interact with epoxy molecules by forming hydrogen bonding with hydrophilic groups. The absorption of water can be attributed to the affinity of the functional groups of the epoxies which having high polarity towards water molecules. Water can be exist in two forms (i) free water which will fill the micro-cavities of the network structure in the composites, and (ii) strong interactions occur within the water molecules and polar group of epoxy resin. Fiber reinforcement in epoxy matrix could reduce the penetration of water in composites <sup>(11)</sup>

In general, diffusant intake decreases as the aluminum-filler content increases, which is reasonable because aluminum filler incorporation in the adhesive decreases the available volume for moisture sorption. <sup>(7)</sup>

The activation energy of all samples is calculated, as shown table {1}.The sample of Al powder/Epoxy has the highest value of activation energy than other samples, see figure (3)

## 5. Conclusions

1) Thermal conductivity of epoxy resin is increased after reinforcing with Al fibers and Al powder in the

normal condition and decreased after immersion in water bath at 40°C for 30 and 60 min .

2) Increasing of temperature of water bath to 50°C led to increase the thermal conductivity to all samples for 30 and 60min of immersion in water.

3) Thermal properties of Al powder is higher than Al fibers because of good interfacial bonding between Al powder and matrix which led to high values of thermal conductivity.

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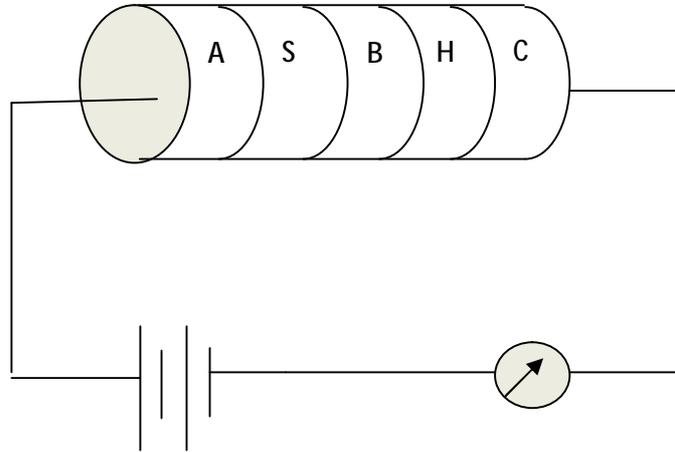
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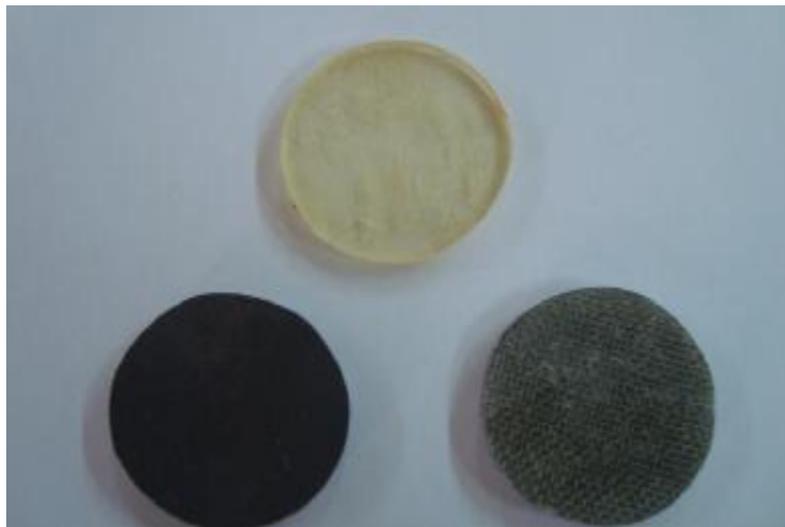
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**Table (1) Shows values of Thermal conductivity and Activation energy for samples**

Samples	Thermal conductivity(w/m.°c)				Activation energy kJ/mole		
	N.C 30°C	Water bath /40°C		Water bath/50°C		30 min	60 min
		30min	60min	30min	60min		
Epoxy	0.394	0.332	0.331	0.507	0.4	38.16	16.54
Epoxy + Al powder	0.866	0.686	0.618	0.873	0.73	140.65	97.19
Epoxy +Al fibers	0.458	0.455	0.404	0.587	0.561	20.52	35.3



**Figure (1) Schematic diagram of Lees Disk**



**Figure (2) Samples of research that coincide (ASTM-D150), (1)Pure Epoxy ,  
(2)Epoxy +Al powder,(3)Epoxy +Al fiber**

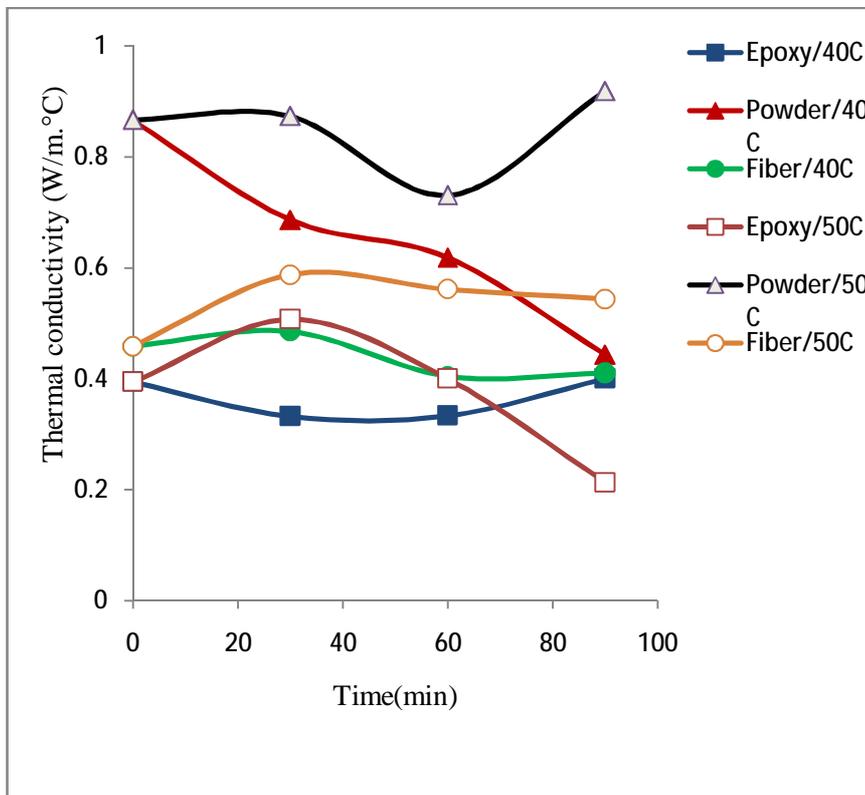


Figure (3) thermal conductivity for all samples at 30, 40, 60, °C for 30 and 60min