

## Prediction and Investigation Particulate Fillers (Aluminum & Alumina) Effect on the Thermal Conductivity of Polymeric Matrix Composite (Polyester)

**Dr. Ahmed A.M.**

Mechanical Engineering Department, University of Technology/ Baghdad  
Email: Aamsaleh60@yahoo.com

**Dr. Ibtihal Al-Namie**

Mechanical Engineering Department, University of Technology/ Baghdad

**Hasan A. Alasady**

Mechanical Engineering Department, University of Technology/ Baghdad

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### ABSTRACT

The main objective of this paper is to study the thermal conductivity of composite materials and the parameters affecting it, then to determine the validity of the best theoretical model matching with experimental results and to predict thermal conductivity of any composite materials with different particle size.

Alumina & aluminum filled thermoset polyester composites are investigated in this study, and their thermal conductivity will be the central focus. Aluminum particle, alumina particle and aluminum fiber (5wt % to 45wt %) were added to polyester matrixes. It was found that both fillers and fiber positively effect on the thermal conductivity of the composite. By using visual basic program, the results show there are three different theoretical models (Maxwell Equation, Lord Rayleigh Equation & Lewis and Nielsen Equation) valid to predict the effective thermal conductivity for reinforced the polyester by Al and Al<sub>2</sub>O<sub>3</sub> powder until 35wt% and 45wt% respectively. In addition, the rule of mixture model is good to predict the effective thermal conductivity (for aluminum fiber parallel with polyester matrix).

**Keywords:** Thermal Conductivity Of Composite, Polymer Matrix Composite (PMC), Filler Size Effect, Al & Al<sub>2</sub>O<sub>3</sub> Particulate.

التنبؤ و التحقق بتأثير الدقائق المائنة (الالومنيوم و الالومينا) على الموصلية الحرارية للمواد المركبة ذات أساس بوليمري (بولي استر)

الخلاصة

الهدف الرئيسي من هذا البحث هو دراسة الموصلية الحرارية للمواد المركبة و العوامل المؤثرة عليها ثم تحديد أي معادلة نظرية افضل تتوافق مع النتائج العملية , و التنبؤ بالموصلية الحرارية للمواد المترابطة مع اختلاف حجم الدقائق .

في هذه الدراسة تم تناول البوليمر المدعم بالالمنيوم و الالومينا و تم التركيز بوجه الخصوص على الموصلية الحرارية . دقائق الالمنيوم , دقائق الالومينا و ألياف الالومينيوم أضيفت إلى المادة الأساس البوليمر بنسب وزنية من ٥% إلى ٤٥%. ان إضافة المادتين المائتين و الليف يؤثر بشكل إيجابي على الموصلية الحرارية . النتائج أظهرت باستخدام برنامج الفجول بيسك هناك ثلاثة موديلات نظرية مختلفة ( معادلة ماكسويل , معادلة لورد ريليه و معادلة لويس نلسن ) كانت متوافقة للتنبؤ بالموصلية الحرارية للبوليمر المدعم بمسحوق الالمنيوم و مسحوق الالومينا إلى النسب الوزنية ٣٥% و ٤٥% على التوالي , و كذلك كانت معادلة قاعدة الخلط جيدة للتنبؤ بالموصلية الحرارية للبوليمر المدعم بألياف الالمنيوم في الاتجاه الطولي .

## INTRODUCTION

Thermally conductive polymer composites can replace metals in many applications. This technology is a substantial improvement since polymers are commonly used due to their thermal properties. The advantages of thermally conductive polymers over metals are reduced density; increased corrosion, oxidation, and chemical resistance; and properties are adjustable to fit the application. The main application for thermally conductive polyester is heat sinks. Other possible benefits are faster injection molding cycle times and improved thermal stability. [1]

A composite material is a macroscopic combination of two or more distinct materials, having a recognizable interface between them [2]. The properties of composite materials are different and improved as compared to constituent materials.

A thermally conductive plastic is ideally suited for heat sink applications, such as lighting ballasts and transformer housings. [3]

Erik H. Weber<sup>[1]</sup> developed a model to predict thermal conductivity of the carbon filled polymer composites and determined if a synergism between fillers exists.

N. Tessier-Doyen et.al. [4] Study the effective thermal conductivity of a two-phase system constituted of alumina particles in a glass matrix. The Maxwell–Eucken expression shown successfully describe the effective thermal conductivity of the glass/alumina mixtures for alumina volume fractions up to 55%.

BhyravMutnuri[5] showed the Addition of 10wt% & 12.5wt% of graphite powder additive in neat vinyl ester resin increased the conductivity by nearly 88%, 170% respectively. He used the graphite powder as a reinforced material with weight ratio 10% & the sizes (44μ, 55 μ, 75 μ & 150 μ) and the result were (0.404, 0.404, 0.346 & 0.335) W/mK respectively. This show that the increasing in the particle size lead to decreasing in the thermal conductivity.

Zhou et.al [6] showed the adding 0–20% Si<sub>3</sub>N<sub>4</sub> filler particles to the polyethylene, lead to increase the thermal conductivity of composite from 0.2 to 1.0W/mK. In addition, the thermal conductivity of composite further enhanced to

1.8W/mK by decreasing the  $\text{Si}_3\text{N}_4$  particle sizes from 35, 3 and 0.2  $\mu\text{m}$ , and using coupling agent, for the composites with higher filler content. Alumina short fibers added to improve the overall composite toughness and strength. Examinations the parameters of the Agari model indicated that smaller  $\text{Si}_3\text{N}_4$  particles could more easily form conductive channels in composites as compared to the larger filler particles.

**RESEARCH OBJECTIVES**

The objectives of this paper were to prepare thermally conductive composites, Characterize and analyses alumina& aluminum filled polyester composites, study the effect of filler sizes on the thermal conductivity of composites samples and determine which is the best theoretical model fit with experimental results to predict thermal conductivity of composite.

**THEORETICAL MODELS**

There are many theories dealing the thermal conductivity of composite materials. In this research, it was chosen theories that are most frequently used.

**Maxwell** [7] derived an equation for effective thermal conductivity of a homogeneous material into which is dispersed many small spherical particles. He selected a characteristic cell and introduced an effective (equivalent homogeneous) medium surrounding this cell as shown in Figure (1).

Maxwell considered the situation where  $n$  small spheres of radius  $r_2$  and conductivity  $k_2$  were embedded within a single larger sphere of radius  $r_1$  and conductivity  $k_1$  such that the temperature fields of neighboring particles are independent of one another.

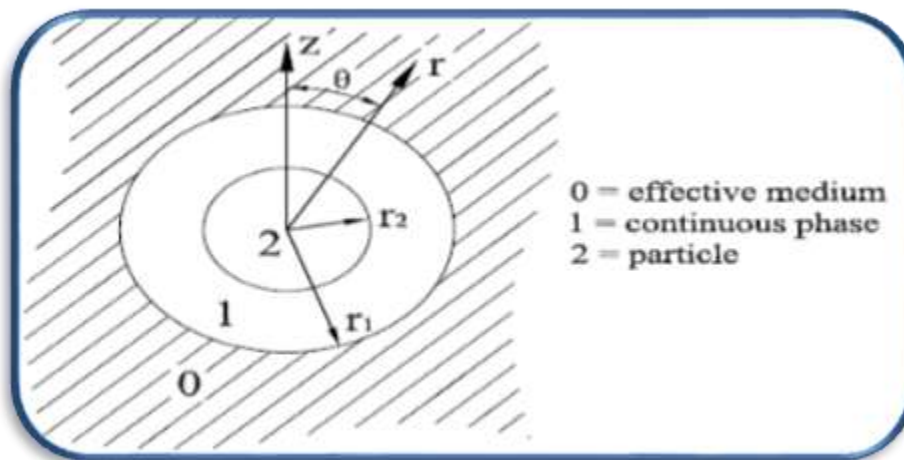


Figure (1) Cell geometry subject to Maxwell's analysis.

The temperature distribution in each component is defined by the independent steady-state diffusion equation,

$$\nabla^2 T_i = \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial T_i}{\partial r} \right) + \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left( \sin \theta \frac{\partial T_i}{\partial \theta} \right) = 0 \quad \dots (1)$$

Where:

- T : temperature
- $\theta$ : the angle
- R: the radius
- i = 0 in the effective medium,
- i = 1 in the continuous phase,
- i = 2 in the solid particle ,

The solution of the independent steady heat conduction equations in the sphere, the continuous phase, and the effective medium subject to appropriate boundary conditions gives the following relationship for effective thermal conductivity:

$$k_e = k_m \frac{2k_m + k_p - 2\phi_p(k_m - k_p)}{2k_m + k_p - \phi_p(k_m - k_p)} \quad \dots (2)$$

Where:

- $k_e$ : Effective thermal conductivity, [W/mK]
- $k_m$ : Thermal conductivity of matrix, [W/mK]
- $k_p$ : Thermal conductivity of particle, [W/mK]
- $\phi_p$ : Volume fraction of particle.

**Lord Rayleigh[8]**

He improved the Maxwell equation by using the principle of multiple pole expansion to take into account the contribution of induced octet pole moments on the field of the neighbor of the central particle. The effective thermal conductivity according to Rayleigh is given by:

$$k_e = k_m \frac{A - 2\phi_p - 0.525B\phi_p^{3.333}}{A + \phi_p - 0.525B\phi_p^{3.333}} \quad \dots (3)$$

Where:

$$A = \frac{2 + k_p}{1 - k_p} \quad \dots (4)$$

$$B = \frac{3 - 3k_p}{4 + 3k_p} \quad \dots (5)$$

**MEREDITH AND TOBIAS**

Meredith and Tobias [9] later improved on the Rayleigh equation. The relationship derived by Meredith and Tobias is given by:

$$k_e = k_m \frac{A - 2\varphi_p + 0.409C\varphi_p^{2.333} - 2.133B\varphi_p^{3.333}}{A + \varphi_p + 0.409C\varphi_p^{2.333} - 0.906\varphi_p^{3.333}} \quad \dots (6)$$

Where:

$$C = \frac{6 + 3k_p}{4 + 3k_p} \quad \dots (7)$$

**Lewis and Nielsen [10]**

They modified the Halpin-Tsai equations for the effective relative shear modulus of a composite to bring it into closer agreement with the experimental data by taking into account the maximum packing fraction of the filler particles. The modified equations for composite systems are:

$$k_e = k_m \frac{1 + AB\varphi_p}{1 - B\psi\varphi_p} \quad \dots (8)$$

Where:

$$B = \frac{\frac{k_p}{k_m} - 1}{\frac{k_p}{k_m} + A} \quad \dots (9)$$

$$\psi = 1 + \varphi_p \left( \frac{1 - \varphi_c}{\varphi_c^2} \right) \quad \dots (10)$$

Where  $\varphi_c$  is the maximum packing fraction of the randomly package heterogeneous media,  $\varphi_c = 0.637$  for spheres. The coefficient A depends upon the geometry and orientation of the dispersed phase, A=1.5.

**THE RULE OF MIXTURE**

For homogeneous fibers of thermal conductivity  $k_f$  embedded in a resin matrix of thermal conductivity  $k_m$ , the thermal conductivity  $k_{ep}$  parallel to the axis of the fiber is given by :[5]

$$k_{ep} = k_f \varphi_f + (1 - \varphi_f) k_m \quad \dots (11)$$

Where:

$k_{ep}$  : Thermal conductivity parallel to the axis of the fiber, [W/mK]

$k_f$  : Thermal conductivity of the fiber, [W/mK]

$k_m$  : Thermal conductivity of the matrix, [W/mK]

$\varphi_f$  : Volume fraction of fiber

**Expert programs for Calculating Effective Thermal Conductivity**

There are many theoretical equations to predict the thermal conductivity; therefore, the visual basic program (version 6) is used to calculate these equations.

In this research the expert programs is prepare to calculate the effective thermal conductivity. The screen face of program is build, select where the input data, calculate command, and out put the result.

**EXPERIMENTAL WORK**

**Materials and sample**

The unsaturated polyester resin used as matrix material (thermal conductivity: 0.4 W/m K). Polyester is Low cost, good handling characteristics, low viscosity, good mechanical strength and Good electrical properties. It can easily machine and carve, as well as molded. Aluminum powder, aluminum fiber and alumina are used as conductive filler material (thermal conductivity: 247 & 39 W/m K respectively).

The weights ratio used (5%, 10%, 15%, 25%, 35% and 45%) figure (2) with three particle sizes for aluminum powder and alumina, also same these weights ratio used with two diameter of aluminum fiber in parallel direction of heat flow.



Figure (2) the samples.

**Equipment & measurement**

The Computer Controlled Heat Conduction Unit is used to measure the thermal conductivity see Figure (3), designed on the principle of Comparative Longitudinal Heat Flow System. The unit was thermally isolated to avoid the heat loss to the surrounding and equipped with several thermocouples at specified locations to measure the temperatures at the core of brass terminals. ASTM: E1225-99 was adopted for Comparative Longitudinal Heat Flow Technique calculations. [11]

The composite samples placed between the two brass terminals of conduction unit. The contact surfaces were coated with conducting paste (supplied along with equipment) to reduce contact resistance and to prevent air gaps. In addition, use equation (12) to calculate the thermal conductivity. [11]

$$k = \frac{Q \cdot \Delta x}{a \cdot \Delta T} \quad \dots (12)$$

Where:

Q: The heat (W),  $\Delta x$  : The thickness of sample.,  $a$  : The area (m<sup>2</sup>)  
 $\Delta T$  : The temperature difference.

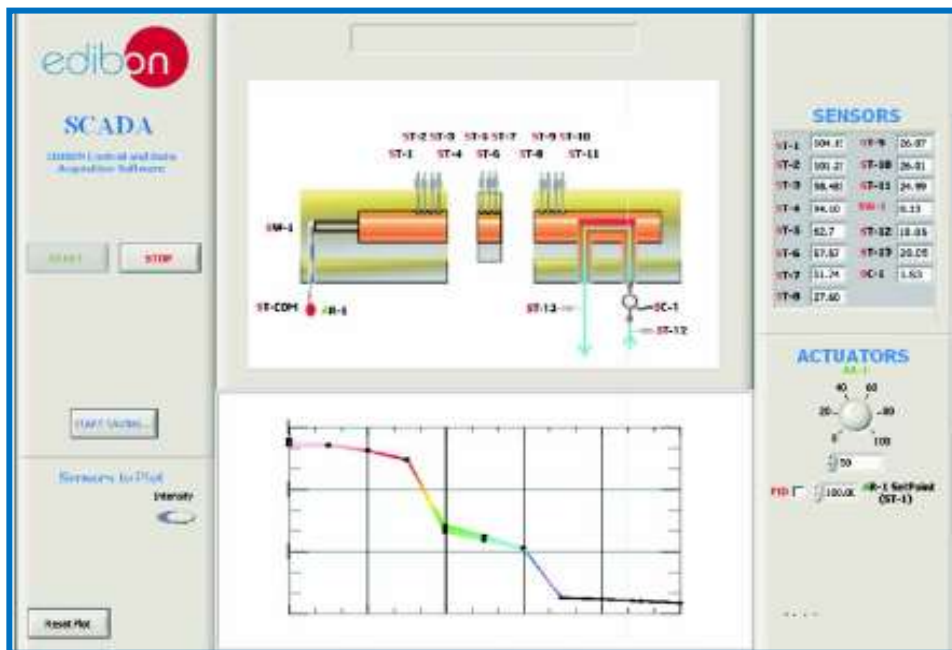


Figure (3) Data from Software of thermal conduction device.

## RESULTS AND DISCUSSION

The Figure (4) shows the effect of weight ratio of aluminum and alumina powder on the thermal conductivity.

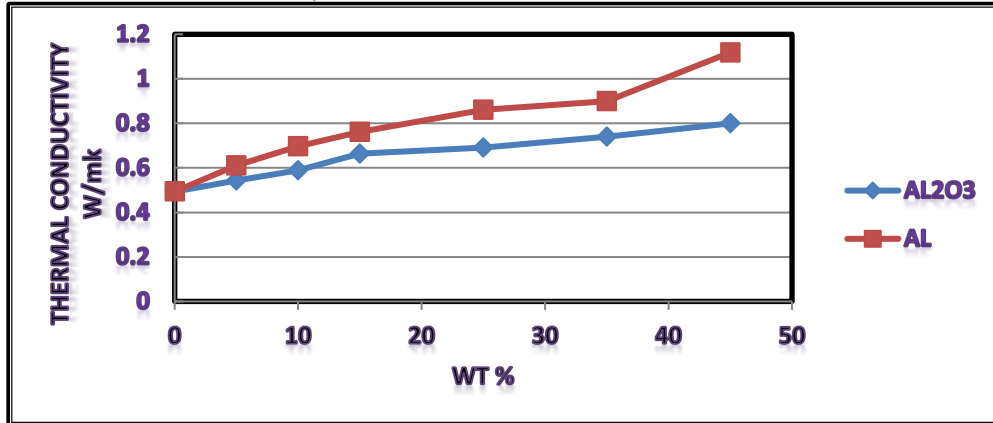


Figure (4) Thermal conductivity for different weight.

#### RATIOS OF AL & AL<sub>2</sub>O<sub>3</sub> PARTICLE COMPOSITE

From Figure (4), it can be seen that the thermal conductivity of composite material increases with the increase of the weight ratio of aluminum particles, because the thermal conductivity of aluminum is high. The heat transfer in aluminum occurs by free electrons and lattice vibration but the heat transfer in the polyester occurs by lattice vibration only, so that the heat finds easy ways with low resistance to transfer through it.

There are three different particle sizes used for aluminum, { (25-90), (90-150) and (150-212)  $\mu\text{m}$  }, and for alumina { below 25  $\mu\text{m}$ , (90-125), (125-150)  $\mu\text{m}$  }.

The magnitude of thermal conductivity of composite material with different particle sizes and different weight ratio of reinforced material for aluminum & alumina is explained in Figures (5&6) respectively.

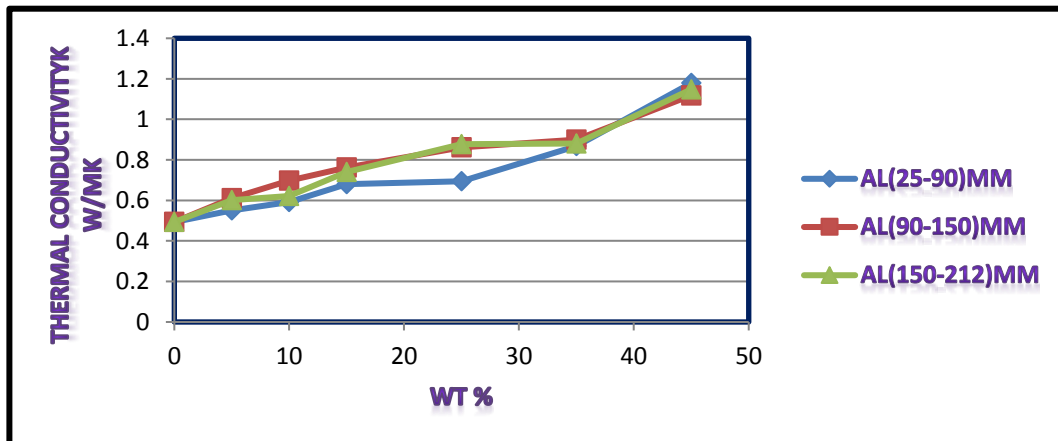




Figure (5) Thermal conductivity with different particle sizes and different weight ratios of Al composite.

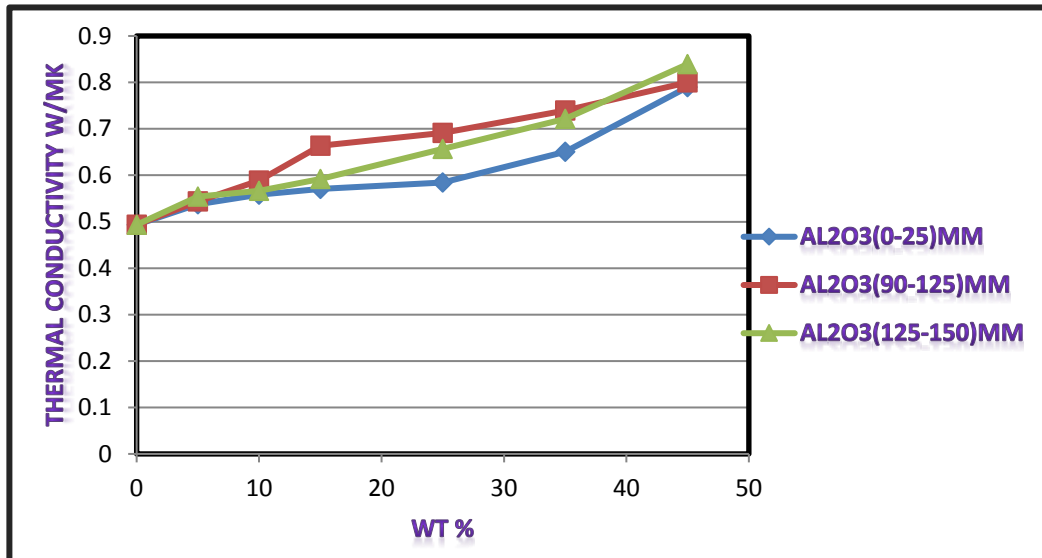


Figure (6) Thermal conductivity with different particle sizes and different weight ratio of Al<sub>2</sub>O<sub>3</sub> particle.

It can be observed From Figures (5&6) that the thermal conductivity of composite material is enhanced with the increase the size of aluminum particle due to the decreases in number of particle so that the contact area decreases and resistance becomes low due to decrease in the overall surface area for the particles same as Zhiguo Li [7].

In other side , if the particle size is very big , it causes very high distance between particles and leads to high resistance between the two particles so that the thermal conductivity become low and the thermal conductivity of composite material enhances with the decrease of the size of aluminum particle due to the increase in number of particles so that the overall surface area of particles increase and thermal conductivity increase, from this explanation, we can understand why the thermal conductivity decrease with increase the particle size of aluminum from (90-150) to (150-212) . This result agreed with result of Bhyrav Mutnuri. [6]

The theoretical results are obtained by using the professional program (VISUAL BASIC PROGRAM) and using four models for particle composite (Maxwell Equation, Lord Rayleigh Equation, Meredith and Tobias Equation & Lewis and Nielsen Equation). Figures (7 & 8) show the results.

Figures (7 and 8) demonstrate the measured thermal conductivity of composite samples and the predicted values of various theoretical models as a function of aluminum (25-90) μm and alumina (below 25) μm weight percentage respectively.

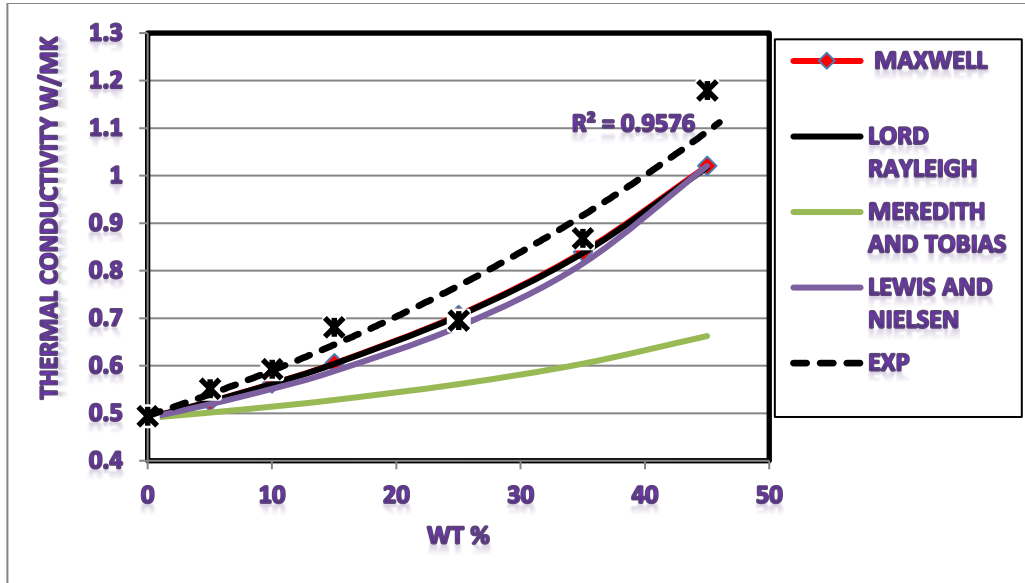


Figure (7) Experimental and theoretical results of effective thermal conductivities of polyester-aluminum.

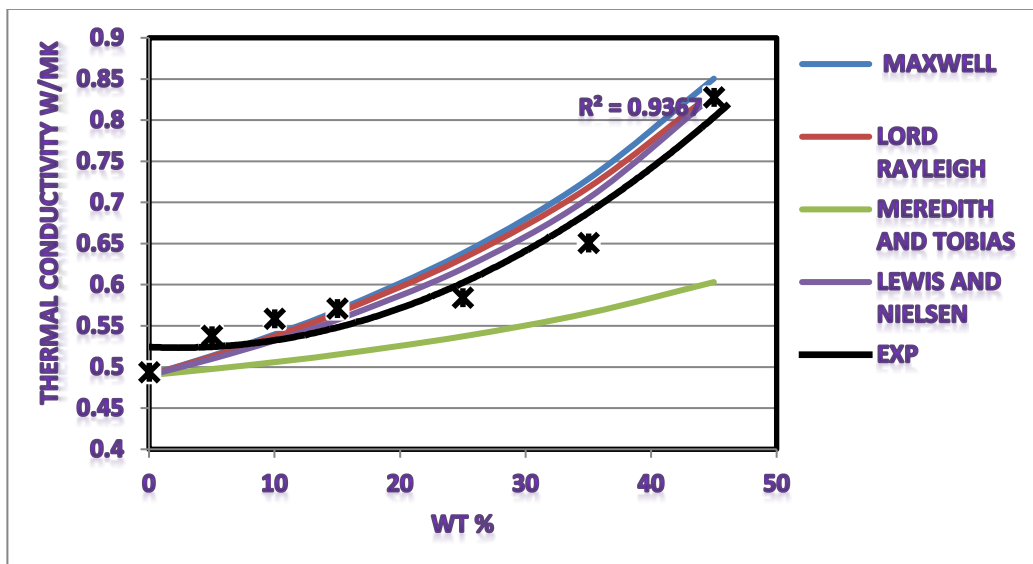


Figure (8) experimental and theoretical results of effective thermal conductivities of polyester-alumina.

It can be observed that the theoretical results lie close to each other at low weight concentrations, however, as the aluminum concentration becomes high, divergences between curves become high. This means that the most of the theoretical models can be conveniently used for the estimation of effective thermal conductivity at low weight fractions, except the theoretical result from Meredith & Tobias.

From Figure (7) , the three models (Maxwell Equation, Lord Rayleigh Equation, & Lewis and Nielsen Equation) are good to predict the thermal conductivity until to the weight fraction 35% (for aluminum particle and polyester). But for higher this weight fraction, the experimental value is higher than the theoretical because increasing the probability of forming thermally conductive chains among filler particles, which may help to improve the heat transferring. Also the Figure (8) showed this three models are good to predict the thermal conductivity to the weight fraction 45% (for alumina particle and polyester).

**THERMAL CONDUCTIVITY OF THE PARALLEL FIBER COMPOSITE**

Figure (9) shows that the thermal conductivity of composite material increases with the increase, the weight percentage of aluminum fiber. This incensement is due to the high thermal conductivity of aluminum. The increase in thermal conductivity is very high because of easy passage to the heat transfer without thermal resistance from the polyester. The increase in thermal conductivity is up to (120.4times of polyester) when adding parallel fiber aluminum with weight ratio (45wt %).

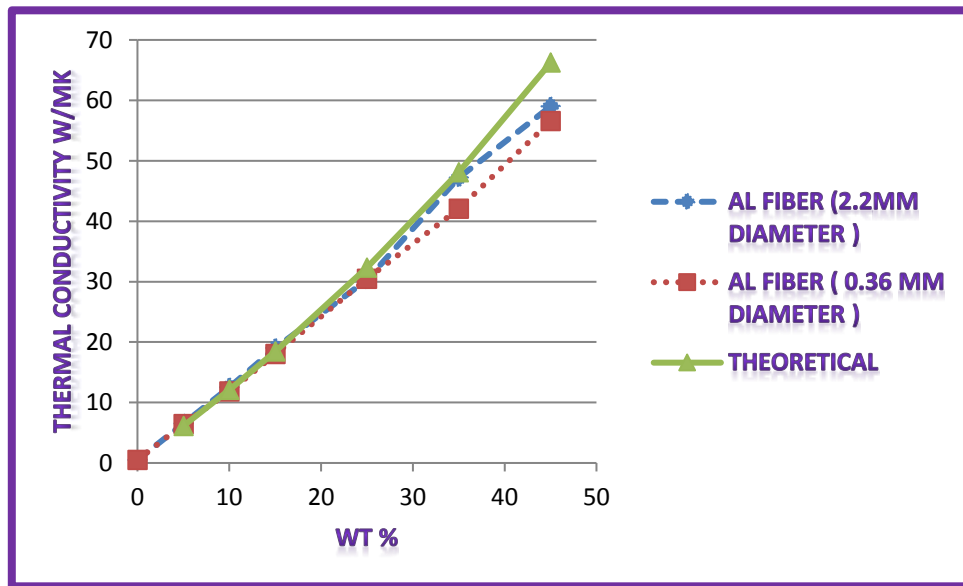


Figure (9) Experimental and theoretical results of thermal conductivities of polyester with parallel aluminum fiber addition.

It is concluded that the rule of mixture model is good to predict the effective thermal conductivity (for aluminum fiber parallel & polyester) as shown in Figure (10). But the effect of the diameter for the aluminum fiber is not significant.

## **CONCLUSIONS**

1. There are very close predicting results to the experimental by using three different methods (Maxwell Equation, Lord Rayleigh Equation, & Lewis and Nielsen Equation) for thermal conductivity with reinforced the polyester by Al and Al<sub>2</sub>O<sub>3</sub> powder until 35wt% and 45wt% respectively. In addition, the rule of mixture model is good to predict the effective thermal conductivity (for aluminum fiber parallel & polyester matrix).
2. The effect of the fiber diameter on the thermal conductivity is not significant.
3. It can be used the polyester with parallel aluminum fiber as heat exchanger plate with low cost, low weight and high resistance to corrosion.

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