



# **Thermal Bridge Development For Uneven Surfaces**

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Sisay Mesele

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Author:	Sisay Mesele
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Supervisor (Arcada):	Mikael Paronen
Commissioned by:	
<p>Abstract:</p> <p>A conductive particulate filled polymer composite is developed by blending Aluminium oxide (<math>Al_2O_3</math>) powder as a conductive filler with various proportions by weight (5,10,15,20,30,40)% wt and Silicone resin as a matrix for thermal bridging application. The material is to be mounted between wall surfaces and flux-meter that was developed at Arcada to measure energy in the form of heat-flux in buildings.</p> <p>Most building surfaces are rough and hold air as thermal insulator and this has a significant effect to create a reading error for the device to give exact value due to the fact that, the rough surface holds air creating a thermal resistance. In order to avoid this thermal insulation by air, <math>k_{air}=0.0027W/m^2.K</math>, the composite material has to fill the rough surface to enhance thermal conductivity between the surfaces.</p> <p>The composition materials were selected based on the application requirement to have characteristics of good thermal conductivity, surface conformability, soft, flexible, self tacky and user friendly to the wall without leaving a stain on the area.</p> <p>Each specimen were tested for thermal conductivity based on experimental measurement and the result revealed as the amount of filler increased thermal conductivity decreased because of increased filler resulted in tough and hard material that incomplete conformation on the wall left air gaps to exist. All samples achieved to conform to surface irregularities, thereby eliminating air voids and improved heat flow through them to conduct heat from the wall surface to the flux meter by illumination of air contained.</p>	
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## Nomenclature

$K$ =Thermal conductivity

$Q$ =heat

$A$ =Area

$Q'$ =heat flux

$L$ =thickness

$t$ =time

$dT$ =change in Temperature

$R_{th}$ = Thermal Resistance

$C_{contact}$  = Contact resistance

TIM= Thermal Interface material

$T_g$ = Glass transition Temperature

$T_m$ = melt transition temperature

$V_f$ = Filler volume fraction

$V_m$ = Matrix volume fraction

$w_f$ = weight fraction of filler

$w_m$ = weight fraction of matrix

$\rho_f$ = density of filler

$\rho_m$ = Density of matrix

$F$ = Packing factor

$K_c$ = thermal conductivity of composite

$K_f$ = filler thermal conductivity

$K_{ef}$ = effective thermal conductivity



# 1 INTRODUCTION

## 1.2 Background

When the heat flux sensor is to be mounted on top of the rough surface of a wall, there is difficult to find the correct measurement of heat energy loss rate, due to the fact that the air trapped in the surface affects by creating a thermal resistance. Since air has one type of thermal insulator with lowest thermal conductivity resulting in lowest conductivity of heat, a thermal interface material is used to fill this air holding spaces on the surfaces. The air gap filler material has to be formulated from a blend of heat conducting materials that can enhance heat transfer from the measurement surface to the measuring device heat flux meter. The composite material is developed from a mixture of two materials, highly soft silicone resin as a base matrix and Aluminum Oxide ( $Al_2O_3$ ) powder as a filler material having high thermal conductivity that can maximize the heat transfer rate from wall to the bottom surface of the flux meter. To minimize the roughness of a wall surface for heat transfer measurement of building surfaces highly soft, flexible and highly conformable silicone elastomeric is chosen as a matrix or base material, the selection is also based on chemical inertness, stain free, naturally tacky and good thermal stability than other polymers are factors for using this material. In addition to this Alumina ( $Al_2O_3$ ) fine powder is incorporated as thermal conductive enhancing filler. The composite material functions as a thermal bridge that interfaces the two solid surfaces, i.e. the heating surface and the bottom surface of the flux meter.

## **1.2 Objective**

The main objective of the thesis is to develop a thermal bridge composite material that can be used as a thermal interface material for the heat flux device found at Arcada to enable to measure the energy transfer through wall surface of a building.

To select and formulate the composition materials based on the required parameters such as thermal conductivity and stability, good filler-matrix integration, flexibility, surface conformation and tackiness as well as user friendly.

The material is used to avoid the air gaps created by the rough surfaces to make smooth and avoid thermal insulation by air.

## **2 LITERATURE SURVEY**

### **2.1 Literature review**

The literature review is to provide background information on the issues to be addressed in this thesis and to emphasize the relevance of the present study. This embraces some related aspects of polymer composites with reference to their thermal conductivity characteristics. The topics include brief review on:

- Thermal properties of composite materials
- Particulate Reinforced Polymer Composites
- Thermal Conductivity of Polymer Composites

#### **2.1.1 On Particulate filled polymer composites:**

Hard particulate fillers consisting of ceramic or metal particles and fiber fillers made of glass are extensively being used these days to dramatically improve the mechanical properties such as wear resistance, even up to three orders of magnitude [20]. Various kinds of polymers and polymer matrix composites reinforced with metal particles have a wide range of industrial applications such as heaters, composites with thermal durability at high temperature [22] etc. These engineering composites are desired due to their low density, high corrosion resistance, ease of fabrication and low cost [10-12]. Similarly for over two decades, ceramic filled polymer composites have been the subject of extensive research. The inclusion of inorganic fillers into polymers for commercial applications is primarily aimed at the cost reduction and stiffness improvement [12, 16]. Along with fiber reinforced composites, the particulate filled composites have been found to perform well in many real operational conditions. Important role in improving electrical, mechanical and thermal properties of the composites is played by silica particles when they are added into a polymer matrix to form a composite, [27, and 28]. Mechanical, thermal and electrical properties of the composites have greatly been affected by the shape, size, volume fraction, and specific surface area of such added particles.

### **2.1.2 Thermal Conductivity of Polymer Composites**

Polymers with particulate filled composite can be classified as commercially available thermal interface material and blend of polymer resin with conductive fillers. In both cases thermal conductivity is enhanced by the particulate fillers since polymers alone cannot transmit heat efficiently; hence polymers have low thermal conductive property. However; some polymers are capable conducting heat through them in low rates that takes time for effective heat transfer. And role of conductive filler particulates are to facilitate heat transfer rate through the composite by forming clusters of conducting site and network by uniform distribution in the polymer matrix [3, 11].

Considerable work has been reported on the subject of heat conductivity in polymers by Hansen and Ho [32] and Tavman [31] etc. But most of these studies were based on the thermal behavior of polymers only and not to their composites. Reports are available in the existing literatures on experimental as well as analytical studies on thermal conductivity of some filled polymer composites [1,29-31]. The fillers most frequently used are Copper particles, short carbon fiber, carbon particles, graphite, Alumina (Aluminum oxide), Aluminum nitrides and zinc particles. The thermal and mechanical properties of copper powder filled poly-ethylene composites are found by Tavman [31] while on thermal properties such as thermal conductivity, thermal diffusivity and specific heat of metal(copper, zinc, iron, and bronze) powder filled HDPE composites in the range of filler content 0–24% by volume were investigated experimentally by Sofian et al. [35]. A moderate increase in thermal conductivity up to 16% of metal powder filler content was observed. The improvement in electrical and thermal conductivity of polymers filled with metal powders was reported by Mamunya et. al [36]. Weidenfeller et al. [37] studied the effect of the interconnectivity of the filler particles and its important role in the thermal conductivity of the composites. They prepared PP samples with different commercially available fillers by extrusion and injection molding using various volume fractions of filler content to systematically vary density and thermal transport properties of these composites.

They measured that the thermal conductivity of the PP has increased from 0.27 up to 2.5 W/mK with 30 vol% talc in the PP matrix, while the same matrix material containing the same volume fraction of copper particles had a thermal conductivity of only 1.25 W/m-K despite the fact that thermal conductivity of copper particles have a value approximately 40 times greater than that of talc particles. Tekce et. al [37] noticed of the shape factor of fillers has a strong influence on thermal conductivity of the composite.

## 2.2 Modes of Heat transfer in materials

The three fundamental types of heat transfer are: Conduction, Convection and Radiation. All engineering surfaces exhibit some level of microscopic roughness. The resistance to heat flow through a contact interface occurs because only a small portion (usually 1-2%) of the nominal surface area is actually in contact [14]. Heat may pass through the interface via three paths: conduction through the contact spots, conduction through the gas present in the gap between the surfaces, and radiation across the gap. Also, radiation does not play a significant role at temperatures below 500°C [18], the working temperature of building surface usually does not exceeding than room temperature .

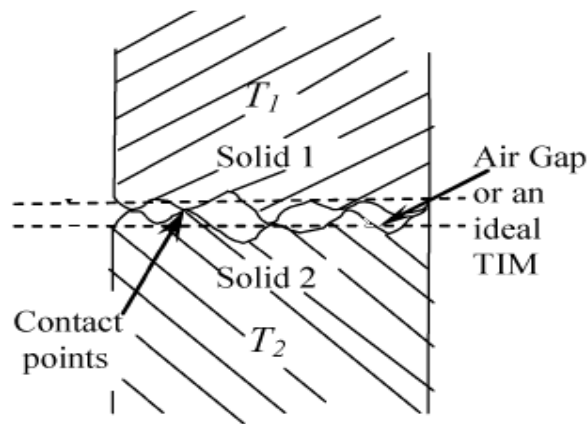


Figure 1. Conductive Thermal Interface material positioned between two solid rough surfaces [19].

### 2.2.1 Conduction

It is the transfer of heat from one part of a body to another part of the same body, or from one body to another in physical contact with it, without appreciable displacement of the particles of the body. Fourier's law is the fundamental differential equation for heat transfer by conduction.

$$\frac{dQ}{dt} = -kA\left(\frac{dT}{dx}\right) \quad (1)$$

Where  $\frac{dQ}{dt}$  (quantity per unit time) is the rate of flow of heat, A is the area at right angles to the direction in which the heat flows, and  $-\frac{dT}{dx}$  is the rate of change of temperature with the distance in the direction of heat, i.e., the temperature gradient. The factor  $k$  is the thermal conductivity, it is characteristic property of the material through which heat is flowing and varies with temperature.

### 2.2.2 Convection

Convection is the transfer of heat from one point to another within a fluid, gas or liquid by the mixing of one portion of the fluid with another. This fluid motion is associated with the fact that, at any instant, large number of molecules are moving collectively or as aggregates. Such motion in the presence of temperature gradient, contributes to heat transfer [9]. Convection is neglected due to the small length scales involved [18], between the thermal interface material and the substrate surface. The convective heat flux  $q'$  is expressed by;

$$q' = h(T_s - T_\infty) \quad (2)$$

Where  $q'$  is heat flux,  $h$  is the convective heat transfer coefficient,  $T_s$  is surface temperature,  $T_\infty$  is temperature of fluid.

### 2.2.3 Radiation

It is the transfer of heat from one body to another, not in contact with it, by means of wave motion through space [8]. And given by;

$$q'_{rad} = \epsilon\sigma(T_s^4 - T_{sur}^4) \quad (3)$$

Where,  $T_s$  is the absolute surface temperature (K) and  $T_{sur}$  is the surrounding

Temperature,  $\epsilon$  is the emissivity constant,  $\sigma$  is the Stefan Boltzmann constant

( $\sigma = 5.67 * 10^{-8} \text{W/m}^2 \cdot \text{K}^4$ )

## 2.3 Heat flux

The heat flux  $q'_x$  ( $\text{W/m}^2$ ) is the heat transfer rate in the  $x$  direction per unit area perpendicular to the direction of heat transfer, and it is proportional to the temperature gradient,  $dT/dx$  [9]. For one dimensional plane wall, having a temperature distribution  $T(x)$ , the rate of equation is expressed as

$$q' = -k \frac{dT}{dx} \quad (4)$$

The measurement of heat flux is most often done by measuring a temperature difference over a piece of material with known thermal conductivity. This method is analogous to a standard way to measure an electric current, where one measures the voltage drop over a known resistor [10].

### 2.3.1 Heat flux sensors (meter)

Heat-flux meters are devices used in both fluid mechanics and thermal engineering to measure local directional heat transfer of heat. They can provide information on the thermodynamic, chemical or mechanical state of a fluid, but they can also be used energy savings analysis building sciences. So the sensor should be mounted on a wall surface through which the heat flux has to be determined [5].

### **2.3.2 Applications in building physics**

In a world ever more concerned with saving energy, studying the thermal properties of buildings has become a growing field of interest. One of the starting points in these studies is the mounting of heat flux sensors on walls in existing buildings or structures built especially for this type of research. In fact building wall surface temperature is mostly set to room temperature, where there is no a tendency to get high surface temperature involvement. In such case, the rate of conductive heat transfer rate is not faster even though the thermal conductivity of the thermal bridge material is higher [10]. Observing Fourier's law of conduction provides good explanation of the principle of heat transfer through the material with a given thickness.

Thermal properties of a wall generally do not change and that it is not always possible to insert the heat flux sensor in the wall, so that it has to be mounted on top of the wall. When the heat flux sensor has to be mounted on top of the wall, one has to take care that the added thermal resistance is not too large [10]. In order to avoid or minimize as much as possible use of thermal bridge (Thermal interface material) between the wall surface and the sensor should be adequately implemented. The performance of this material is selected based on surface conformability to fill air gaps that can increase thermal contact resistance to reduce and facilitate heat transfer from the wall through the sensor device. In addition to this the selected material should be able to conduct heat as desirable.

## **2.4 Factor affecting heat flux measurement on uneven surfaces**

Surface texture, as it is discussed above surface roughness creates a thermal barrier by holding air, which it thermal insulator. It is known that most building are made from concrete and other construction materials that the outer surface are not smooth and finishing is usually difficult and even "real" surfaces are not perfectly smooth and flat. When two solids are joined as fig [1] illustrates, imperfect joints (interfaces) are formed. The imperfection can only be avoided by interfacing a material with a better thermal conductivity in the contacting solids avoids the unevenness and facilitates thermal transfer.



## 3 MATERIAL SELECTION

### 3.1 Overview of material selection

The vast availability of material existence with several characteristics is the main challenge in material development and compositions world. Existing wide range of materials as well as variety of requirements in the design procedure are roots of the difficulty of the selection procedures. Suitable material selection based on the requirement for the optimum application of heat transfer is the key idea of this paper. It is known that materials of high thermal conductivity are needed for the conduction of heat for the purpose of heating or cooling. However; it needs through look to choose the compositions i.e. Matrix and filler to fulfill the required task and to prepare a composite material.

Several types of thermally conductive materials can be used to eliminate air gaps from rough surfaces such as, thermal interface, including greases, reactive compounds, elastomers and pressure sensitive adhesive films. All are designed to conform to surface irregularities, thereby eliminating air voids and improving heat flow through the thermal interface [3, 14, and 15].

As it was mentioned the aim in this thesis is to find material composition in both polymer and filler particulates that enhance thermal conductivity. The final product likely to have the following important qualities: possible higher thermal conductivity, Easily deformed by small contact pressure to contact all uneven areas of the surface, so that eliminating  $R_{\text{contact}}$ , minimal thickness to help maximize the rate of heat transfer, non-toxic to avoid any hazards to the user, maintain performance indefinitely, and manufacturing friendly (easy to apply and remove).

A blend of two materials to create the required composite structure is studied in order to understand and fulfill the desired properties. In this case, since several polymeric materials exist with most of them having unique and diverse properties and the rest similar characteristics, it is exhaustive in order to identify the appropriate polymer matrix based on the properties they have for the required application. In addition to this, most poly-

meric material properties are influenced by external factors such as temperature, light, pressure and also their chemical compositions that determine properties significantly.

Secondly, flexible property of the polymer matrix is a property needed. This means, the polymer that acts as a matrix should be able to be reworked with no effort to the desired shapes, better elasticity, thermal stability and malleability are the characteristics required for the application.

It is also necessary to put into consideration thermal properties of polymeric material based on their intermolecular orientation. The rate of heat transport in and through polymers is of great importance. For good thermal insulation the thermal conductivity has to be low, on the contrary for conduction purpose it has to be higher [13].

## 3.2 Thermal properties of Materials

### 3.2.1 Thermal conductivity

Thermal conductivity is an inherent or absolute property of the material. The time rate of heat flow through a unit area producing a unit temperature difference across a unit thickness.

$$k = \frac{dq * L}{A * \Delta T} \quad (5)$$

Where,  $k$  is the thermal conductivity of the material,  $dq$  rate of change in heat transfer ( $\text{W}/\text{m}^2 \cdot \text{K}$ ),  $L$  is material thickness (m),  $A$  is heat transfer area ( $\text{m}^2$ ) and  $dT$  is change in temperature ( $^{\circ}\text{K}$ )

It is the intrinsic property of materials to conduct heat. Normally one-dimensionality, steady state operation, homogeneity of the material are given as simplifying premises. The thermal conductivity of any homogeneous material is independent from its thickness. This is not the case with non-homogeneous materials such as elastomers or composites being reinforced by engineering plastics or glass fibers [14, 19].

### 3.2.2 Thermal Expansion

Upon heating, some polymeric materials experience very large thermal expansions on heating as indicated by the values that range from  $\sim 50 * 10^{-6}$  to  $400 * 10^{-6}/^{\circ}\text{C}$ . Linear and branched polymers show highest values because the secondary intramolecular bonds are weak, and moreover, there is a minimum of cross linking. The magnitude of expansion coefficient diminishes with increased cross linking. The lowest values are found in thermosetting network polymers, e.g. phenol-formal-dehyde in which the bonding is almost entirely covalent [41].

### 3.2.3 Thermal resistance

The opposition to the flow of heat through a unit area of material across an undefined thickness. The thermal resistance through the material itself  $R_{\text{th Material}}$  is an internal resistance and is defined after Fourier series development of conduction:

$$R_{\text{th Material}} = \Delta T / Q \text{ or } L / (k A) \quad (6)$$

Factors that affect thermal resistance of the conducting material are material thickness, the thermal conductivity and surface area [15, 17].

The thermal resistance of soft materials such as compliant elastomers and gap fillers are subjected to pressure to achieve surface gap conformations.

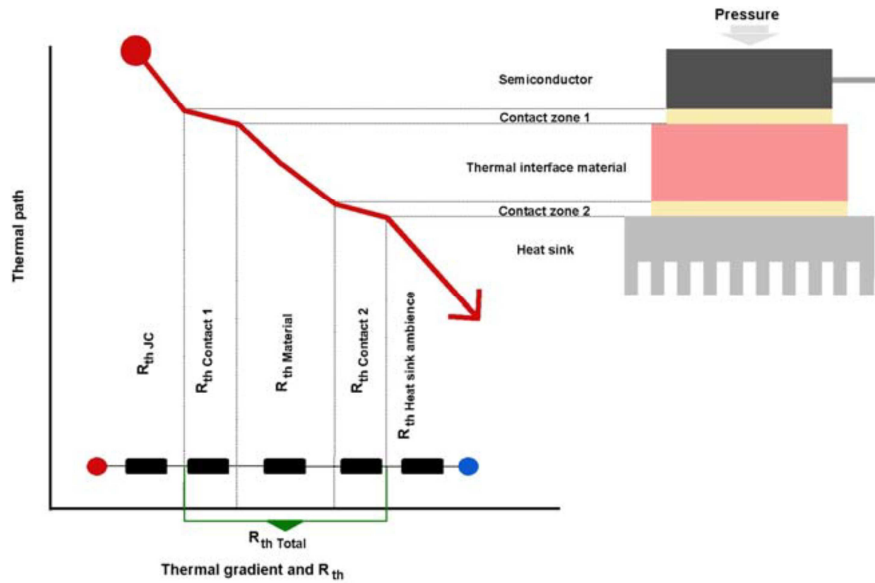


Figure 2 Thermal gradients along the thermal path [14].

In addition to the internal material resistance, the temperature gradients on both contact surfaces are decisive [4, 14]. They affect two contact resistances  $R_{th\ Contact\ 1}$  and  $R_{th\ Contact\ 2}$ . So the overall thermal resistance can be defined as:

$$R_{th\ Total} = R_{th\ Material} + R_{th\ Contact\ 1} + R_{th\ Contact\ 2}$$

Here the equation can be modified based on the assumption that, using thermal interface material can avoid the contact gap by creating adhesive character in both contacting surfaces firmly and both will be ignored. So thermal resistances reduced to:

$$R_{total} = R_{th\ material}$$

Thermal contact resistances are mainly caused by incomplete fitting and air interstitials affecting the border areas with  $k_{Air} = 0,027\ W/m^2\text{-K}$  at the acting as thermal insulators [14,19]. Figure below shows the principle of linear temperature drop from surface temperature where heat dissipation occurs through thermal bridge to heat sink, i.e, and flux meter.

### 3.2.4 Thermal Transmittance

Thermal transmittances commonly known as the U-value, is the measure of the rate of heat loss of a building component. The U-value is calculated from the reciprocal of the combined thermal resistances of the materials in the element, air spaces and surfaces, air gaps. It is expressed as watt per meter, per degree Kelvin ( $\text{w/m}^2\text{-k}$ ), and defined by expression analogues to Newton's law of cooling.

$$Q=UA\Delta T \quad (7)$$

### 3.2.5 Specific Heat Capacity

*Heat capacity* of a solid material is a property that is indicative of material's ability to absorb heat from the external surroundings; it represents the amount of energy per unit mass required to produce a unit temperature rise. Mathematically, heat capacity ( $C$ ) of a body is defined as

$$dQ = Cm\Delta T \quad (8)$$

Where  $dQ$  refers to the quantity of heat transferred to the body and  $\Delta T$  is the rise in temperature. Ordinarily, specific heat capacity( $C$ ) is specified per mass of material (e.g.,  $\text{J/g-}^\circ\text{C}$ , or  $\text{J/kg-}^\circ\text{C}$ ).

### **3.3 Polymer Blends**

There is a trend toward specialization in the polymer products industry. Since the industry is expanding globally, a sufficient market is available for polymer blend products. Blending is a convenient route to time-efficient and cost-effective upgrading of commodity and engineering resins and to tailoring these resins to specific performance profiles for the desired application [20].

Thermal and rheological behavior is affected by addition of fillers, and thus these have impact on processing. Adding conductive fillers such as glass fiber or Aluminum oxide increases the thermal conductivity, but only slightly affects the specific heat. The presence of fillers adds new phases to the system (the reinforcement and the sizing agent) and creates several new interfaces: between the new phases and each of the phases already present in the system. Each of these interfaces has to be managed. Proper interactions have to be ensured between the different phases to obtain the desired product performance [20, 11].

In industrial fabrication, polymers blending with other non polymeric materials are composites. The blending process and types of matrix filler reinforcement influences the characterization of material for desired application; hence for the purpose of thermal conductivity enhancement it can be categorized as thermal conductive composites.

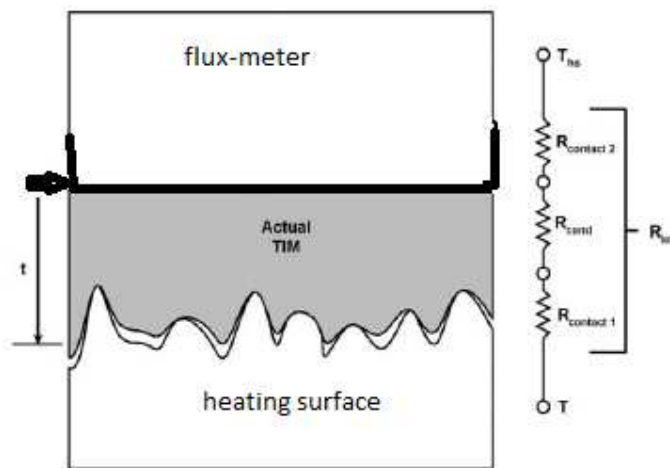
### **3.4 Thermal Interface Materials and Conductive Composites**

TIM are thermally conductive materials, which are applied to increase thermal conductance across joined or parallel solid surfaces in order to increase thermal transfer efficiency. Using a TIM of high thermal conductivity that can conform to the imperfect surface features of the mating surfaces.

Any interstitial material that can fill the gap between contacting surfaces, and whose thermal conductivity exceeds that of air will decrease the contact resistance [6]. The working principle of thermal bridge can be described as, when two component surfaces are brought together, a very few percent of the surfaces make physical contact. As much as large percent of the surfaces are separated by a layer of interstitial air.

Some heat is conducted through the physical contact points, but much more has to transfer through the air gaps. Since air is a poor conductor of heat, it should be replaced by a more conductive material to increase the joint conductivity and thus improve heat flow across the thermal interface [3].

Therefore, by use of thermally conductive interface materials, both the contact resistance and the overall resistance are lowered. The better the material complies with both surfaces, the less air interstitials there are. It minimizes the reading error that occurs during energy measurements [14, 15]. The overall target is to lower the thermal conduct resistance created by air in the rough surface and increase the maximum heat conduction between the surface and the device.. However, choosing a material with higher thermal conductivity dominates for such application in terms of heat transfer compliance.



**Figure 3 Thermal interface materials in place with flux meter**

As shown in Fig. the interface resistance is composed of three resistances in series, assuming little effect of thermal resistance between the thermal bridge and heat flux meter  $R_{\text{contact } 2}$  will be ignored in the analysis.  $R_{\text{contact } 1}$  is the contact resistance between the TIM and the wall surface,  $R_{\text{cond}}$  is the conduction resistance across the thickness of the TIM,  $R_{\text{contact } 2}$  is the contact resistance between the TIM and the heat flux meter base

plate and also be ignored in the analysis,  $A$  is the surface area of the wall component across which heat transfer occurs, and  $t$  is the thickness of the TIM.

The heat flux measurement is set according to the figure 3 shown, where the actual thermal bridge (TIM) material is mounted on the surface of the wall to fully conform on roughness, to achieve this, mounting pressure is needed to fill the thermal bridge on the void surface. It can be applied either by hand or by other means that can assist handling the material without altering the material temperature that may lead to measurement errors. Increasing contact pressure can also decrease the conduction resistance,  $R_{\text{cond}}$ , of the interface material itself, by reducing its thickness. The pressure required to reduce the thermal interface resistance of a given TIM is a function of its compressibility and surface properties as well as the surface properties of the mating surfaces [4].

### **3.5 Particulate Filled Polymer composites**

Modification of organic polymers through the incorporation of additives yields, multi-phase systems containing the additive embedded in a continuous polymeric matrix. The resulting mixtures are characterized by unique microstructures or macrostructures that are responsible for their properties [12].

Polymer composite filled with metal are of interest for many fields of Engineering. The interest arises from the fact that the thermal and electrical characteristics of such composite are close to the properties of metals [1]. The transfer conditions of heat flow determine the thermal conductivity level in heterogeneous polymer filler system, in which the conductive phase is formed by dispersed metallic or carbon filler. The influence of the type of polymer matrix and filler interaction has a great property characterization effect [13]



### **3.5.1 Types and Composition of Polymer Composites**

Polymer composites are mixtures of polymers with inorganic or organic additives having certain geometries (fibers, flakes, spheres, and particulates). Thus, they consist of two or more components and two or more phases. Additives for polymer composites have been variously classified as reinforcements, fillers, or reinforcing fillers. Reinforcements, being much stiffer and stronger than the polymer, usually increase its modulus and strength. Thus, mechanical property modification may be considered as their primary function, although their presence may significantly affect thermal expansion, transparency, thermal stability, and so on [6].

Continuous composites contain long fiber or ribbon reinforcements mostly in thermosetting matrices; when prearranged in certain geometric patterns, they may become the major component of the composite (they can constitute as much as 70% by volume in oriented composites) [10].

In discontinuous composites, the directional reinforcing agents (short fibers or flakes) are arranged in the composite in different orientations and multiple geometric patterns, which are dictated by the selected processing and shaping methods [11]. In order to be able to select the material composition according to the needed use, both the matrix and the filler material should have characteristics that are suitable for the desired application

### 3.5.2 Polymer matrix properties and selection

The characteristics required for the base matrix material is the most influential component in developing polymer matrix composite for this specific task. Therefore in order to select the best alternative, the objective is to find a polymer or polymeric derivative material that has a flexible, soft, easy deformability, good elasticity, non toxic property. Additionally conformability to roughness is the main characteristics desired, due to the fact that, identifying a suitable polymer and conductive filler that can avoid the volume of uneven surface of a wall without the requirement of additional force to mount on the wall surfaces. The filling of polymer with metallic particles results in an increase of both thermal and electrical conductivity of composites obtained [1]. So that selection and formulation of suitable matrix (polymer) for such application is important part of this work.

Adhesive matrix is used to form a mechanical bond at an interconnection. Thermoplastic adhesives are rigid materials at temperatures below the glass transition temperature ( $T_g$ ) of a polymer. Above the  $T_g$ , polymers exhibit flow characteristics [17]. Thus, the  $T_g$  must be sufficiently high to avoid polymer flow during the application conditions [11,6], but the working surface area is at room temperature i.e, there happens to be steady state heat transfer to the surrounding environment, and polymers depend on their  $T_m$  and  $T_g$  to initiate their characteristics are not useful for this purpose.

Thermosetting adhesives, such as epoxies and silicones, form a three dimensional cross-linked structure when cured under specific conditions. Cure techniques include heat, UV light, and added catalysts. As a result of the cure reaction which is irreversible, the initial uncross-linked material is transformed into a rigid solid. The thermosetting are stable at high temperatures and, more importantly, provides a low contact resistance. This results from a compressive force that maintains the conductive particles in intimate contact after the cure.

That is, the shrinking caused by the cure reaction achieves a low contact resistance with long time stability. The ability to maintain strength at high temperature and robust adhesive bonds are the principal advantages of these materials. However, because the cure reaction is not reversible, rework or repair of interconnections is not an option [10,17].

Silicone elastomer, and other similar elastomers can be available or developed in a sheet form with ceramic particles as conductive filler and or fibers imbedded in them to increase the thermal conductivity. These are especially good for temporary assemblies or setups since TIM removal is simple and often reusable. There are also gap filling versions of these sheets such as those made by Fujitsu electronics manufacturer. These TIMs require a moderate to high compressive force to work effectively. Thermal conductivity ranges from 0.5 W/m-K to 3.5 W/m-K [17].

### **3.5.3 Particulate Filler materials**

Fillers are different from additives, in the sense that they are only mixed with the polymer matrix to impart specific physical properties instead of chemical properties such as mechanical, strength, electrical conductivity or dielectric properties, thermal insulation and they never take part in the macromolecule chain. Because fillers together with the polymer matrix are considered as composite materials. Fillers can constitute either a major or a minor part of a composite [12].

Fibers and whiskers are not discussed here because they are generally regarded as reinforcements, not fillers, although a majority of the fillers have reinforcing effects. Also, fillers and additives that primarily modify or impart electromagnetic properties, such as electrical conductivity, are not discussed.

### **3.5.4 Characteristics of Particulate fillers in composites**

In formulating filler polymer composite, the developer should consider important properties and qualities the filler must have before the blending process. This is because filler can define the final properties of composite product that should be needed. It is necessary to raise and answer such questions, what property benefits are being sought, deleterious changes that may occur and how can they be tolerated, how easy is the filler to handle and how might affect processing and so on[11].

Understanding the chemistry of compounding the filler into the matrix determines the interaction between polymer and filler results the predefined product, while the chemical nature of the filler has little direct importance to its use in composites. They can be divided in to two categories inert and reactive, while the later can reacts with the environment.

The other factor that affects the behavior of a composite is surface chemistry and surface interaction because polymers have a very much higher (20-30 times) thermal expansion coefficient than mineral and metallic fillers. Thus, in many well-dispersed, hot processed composites, a compressive stress develops as the polymer cools leading to an intimate interaction between matrix and filler [12].

The overall value of filler is a function of intrinsic material characteristics, such as particle size, particle shape, and chemical composition; of process-dependent factors, such as particle-size distribution, surface chemistry, particle agglomeration, and bulk density; and of cost [10] determine the structural properties of composite material.

### **3.5.5 Filler Loading**

The amount of filler in a filled polymer is termed the loading and is expressed quantitatively although the quantitative measures vary from industry to industry. In plastics and rubber industries, filler loading is formulated according to parts of filler used per 100 parts of polymer, weight percent (wt %), or volume percent (vol%) [11]. The optimal loading of fillers in a polymer is a balance between property enhancement such as thermal and other physical and processing and material cost over the filler loading range. [12].

### 3.5.6 Density and void fraction:

The average mass per unit volume of the individual particle is the true density or specific gravity of the filler. It is used to calculate the volume fraction of fillers. Densities of finely divided, porous, and irregular fillers are typically measured by a gas pycnometer that ensures all pores and crevices of filler agglomerates are penetrated. Apparent, or bulk, filler density refers to the total amount of volume occupied by a given mass of dry fillers and it includes the void volumes in the filler aggregates and agglomerates. Bulk density is used in weighting fillers during filler purchasing, shipping, and storage [10].

The theoretical density of composite materials in terms of weight fraction can be obtained by equations given by Agarwal and Broutman [32].

$$\rho_c = \frac{1}{\left[\frac{wf}{\rho_f} + \frac{wm}{\rho_m}\right]} \quad (9)$$

Where,  $w$  and  $\rho_c$  represent the weight fraction and density of the composite respectively. The suffix  $f$ ,  $m$  and  $c$  stand for the filler, matrix and the composite materials respectively.

### 3.5.7 Mixing and Dispersion

In preparation of filled polymer composites, the process of uniformly distributing fillers is most important process. Component mixing consists of three basic elements: incorporation, distribution, and dispersion. These occur simultaneously throughout the mixing cycle. Incorporation, which predominates the early stage of mixing, involves blending of separate ingredients into a coherent, although still inhomogeneous, mass. In the distribution step, homogenizing occurs in which the ingredient become randomly distributed throughout the polymer matrix [12]. In industrial production all can be done by electrical drive machine, the so called mixers and homogenizers. Since lab scale is carried out in this task all the blending mechanism are done by manual mixing of ingredients, that involves thorough mixing components until uniform dispersion of filler particle is achieved in the silicone resin.

### 3.5.8 Effects of Filler on composite structure

Filler particulate can significantly affect the structure of the matrix polymer itself and the properties of the final composite. Matrix modification can occur and the effect on properties can be at least as important as that arising from the interfacial bonding effects. Ways in which particulate phase may affect polymer structure are; Molecular weight reduction during processing and crosslink modifications due to interference with curing processes, adsorption of polar, low molecular weight species such as surfactants, plasticizers, stabilizers and antioxidants. [13].

Rheology and Process ability effect; the presence of fillers affects the flow characteristics of the filled polymers. The immediate effect of filler is to increase viscosity, interfere with the polymer flow pattern in a given process [11, 23]. Therefore it is necessary to study the characteristics of filler and the interaction with a base polymer resin to make a uniform dispersion, and so prevents cluster formation of particle concentration in one place, it also upgrades the quality of the composites especially designed for thermal performance applications [16]; hence it is a preliminary and important to study the behavior of both filler and polymer aggregates in order to achieve the most out of the performance required.

Other important filler effect to consider; since polymers generally have a much larger thermal expansion coefficient than most rigid fillers, there is a significant mismatch in thermal expansion in filled polymer composites. This mismatch could lead to generation of thermal stresses around filler particles during fabrication and, most severely, induce micro cracks at the filler interface that could lead to premature failure of the filled polymer [25].

### 3.6 Particle packing

The packing principles are of great value in preparing filler size distributions capable of being processed at high loadings in polymeric composite systems. The packing behavior of particles in a polymer matrix determines at what loading particle/particle effects become important and is a critical factor in the understanding and design of polymer composites, especially when highly filled systems are involved.

One of the most important characteristics of the filler-polymer composite is the filler packing factor  $F$  [1]. The value of  $F$  depends on the particle shape and on the possibility of the skeleton or chained structure formation.  $F$  is a limit of system filling and equal to the highest possible filler volume fraction at a given type of packing.

$$F = \frac{v_f}{(v_f + v_m)} \quad (10)$$

Where  $V_f$  is the volume occupied by the filler particles at the highest possible filler fraction and  $V_m$  the volume occupied by the polymer matrix (space among filler particles)

The maximum packing fraction,  $F_f$ , is a particularly useful concept. This is the maximum volume fraction of particulate that can be incorporated before a continuous network is developed and voids begin to appear in the composite. In addition to indicating the maximum practical loadings obtainable,  $F_f$  is also useful in understanding and describing the effect of filler loading on composite properties. Many properties change rapidly as one approaches  $F_f$ , almost in a percolation way.

### **3.6.1 Particle Packing Theory**

The aims of particle packing theory are to predict the maximum volume fraction that can be obtained under a given set of circumstances and the structure of the particle assembly at this point. Different ways of packing the same collection of particles, each one resulting in a different maximum volume fraction. Ordered packing, for instance, is the circumstance where there is a perfect particle packing with no free space occurs. However; in random packing, the situation where pieces are put in container and shaken and it is unlikely that they will ever discover this perfectly packed structure. Nevertheless, it will be found that, providing there are enough particles, a certain packed density can be reproducibly obtained implying that some statistically balanced structure is achievable[13,22].

Random packing is more relevant to particulate-filled composites, is important in understanding the application of packing theory, and two classes of random packing are recognized, loose and dense random packing. Loose packing refers to the sort of packing obtained when particles are randomly assembled under conditions where they cannot easily move past each other, while in dense random packing, conditions are such that movement is possible [11,26].

### **3.7 Additives**

Polymers can incorporate in their structures additives that enhance some specific properties of the polymer. However; most of them are chemical compounds that affect chemical properties, and more common types of additive are, dyes, plasticizers, solvents, stabilizers, lubricants and antioxidants. The primary reasons for using additives are, Property modification or enhancement, overall cost reduction, Improving and controlling of processing characteristics [6]. They are also added to polymers to characterize and enhance physical, and chemical properties according to the required performance of a given composite product [10,32].



### 3.8 Theoretical Background of Conductive Polymer Composites

Polymeric matrices have significant amount of thermal and electrical properties and thus are thermal and electrical insulators. The conductive fillers provide both thermal and electrical properties and the polymeric matrix provides both thermal and mechanical properties. Therefore, thermal, electrical and mechanical properties are provided by different components, unlike that of metallic solders that provide all these properties [17, 23].

When a polymer matrix having a conductivity  $K_m$  is filled with dispersed filler having a conductivity  $K_f$ , the composite prepared gains a conductivity value  $K_c$ . when the volume filler fraction  $V_f$  reaches a critical value  $V_c$  (so called percolation threshold), an infinite conductive cluster (IC) is formed and consequently, the composite becomes conductive [1]. As the filler concentration increases from  $V_c$  to the filling limit  $F$ , the value of  $K_c$  increases rapidly over several order of magnitude, from the value  $K_c$  at the percolation threshold, the conductivity change is negligible and the conductivity of the composite is equal to the polymer conductivity  $K_c$  or slightly higher.

#### 3.8.1 Theoretical Models of Thermal Conductivity in Composites

Due to the fact that Polymers have lower thermal conductivity compared to metals and many inorganic materials the thermal conductivity can be enhanced by filling a high thermal conductive filler particulates, or metallic powder such as Aluminum oxide, silver, Zinc or graphite [12, 25].

Theoretical models have been proposed to predict the effective thermal conductivity of two-component composites. For two-component composites, the simplest consideration would be that the components are arranged in either parallel or series with respect to heat flow, which give the upper or lower limits of the effective thermal conductivity of composites. For the components in parallel,

$$K_{ef} = (1 - V_f)K_m + V_fK_f \quad (11)$$

and for the components in series,

$$\frac{1}{k_s} = \frac{1 - V_f}{k_m} + \frac{V_f}{k_f} \quad (12)$$

For composites with irregularly dispersed fillers in a continuous matrix, Maxwell [18], studied their effective thermal conductivity. The resulting effective thermal conductivity of this system is expressed as,

$$k_e = k_m \frac{2k_m + k_f + 2V_f(k_f - k_m)}{2k_m + k_f - V_f(k_f - k_m)} \quad (13)$$

Where  $k_e$  is the effective thermal conductivity of the composite,  $k_m$  is the thermal conductivity of the matrix,  $k_f$  is the thermal conductivity of the fillers and  $V_f$  is the volume fraction of the fillers.

Thus, once the mixing proportion of both filler and resin are known, according Maxwell correlation, the thermal conductivity of the emerging composite material can also easily be determined. The thermal conductivities of filled polymers typically lie in between the limits set by rule of mixtures.

## **4 METHODOLOGY**

This chapter describes the materials and methods used for the processing of the composites under investigation. It presents the details of the characterization and thermal conductivity tests which the composite samples are subjected to. In addition to this thermal conductivity of the prepared Aluminum oxide filled composite samples are tested for thermal conductivity and compared the theoretical assumptions with the experimental values obtained.

### **4.1 Materials**

#### **4.1.1 Resin**

Silicone elastomers are elastic substances which contain linear silicone polymers cross linked in a 3-dimensional network. In most cases this network also contains filler which acts as a reinforcing agent or as an additive for certain mechanical, chemical or physical properties. In general all silicones (usually we refer to silicones as polydimethyl siloxanes) are noted for their high thermal stability, biocompatibility, hydrophobic nature, electrical and release properties. When silicones are cross linked to form a silicone rubber their characteristic properties are still prevalent [7]. The nomenclature classifies silicone elastomers by their curing mechanism and curing conditions. They are essentially divided into two groups of materials, i.e., room temperature vulcanizing (RTV) and high temperature vulcanizing (HTV). RTV systems are able to cure at room temperature and HTV systems at temperatures well above 100 °C. A number in the name indicates the number of components that upon mixing will form a curable composition, e.g., RTV-2. HTV rubbers are mainly so-called solid silicone rubbers that are not suitable for desired application. They have a very high viscosity in the uncured state and appear as solids. This behavior has also led to the creation of the term 'High Consistency Rubber' [7, 26].

Low viscosity silicone resin from Wacker, was used as a matrix material. Known by its trade name Silicone Elastosil M4601 A, Pourable, addition-curing, two-component silicone rubber that vulcanizes at room temperature (RTV-2) and the corresponding hardener Elastosil M4601B (platinum Catalyzed) are mixed in a ratio of 9:1 by weight as recommended by the manufacturer [26].

It is chosen primarily because of the above mentioned reasons as well as it happens commonly used polymer for it has got a thermal stability and conductivity in comparison to other polymeric material, about 0.2 W/m-K, also depending on the curing process and the amount of catalyst used, tacky (adhesive property), easily deformable to the desired shape, flexible, elastic and chemically inert compound can be achieved, if property characterization is applied to fulfill the desired application.

<b>Property</b>	<b>Inspection Method</b>	<b>Value</b>
Elongation at break	ISO 37	700 %
Hardness Shore A	ISO 868	28
Mix ratio at	A : B	9 : 1
Tensile strength	ISO 37	6.5 N/mm <sup>2</sup>
Viscosity at 23 °C, after stirring	ISO 3219	15000 mPa s
Viscosity at 23 °C	ISO 3219	10000 mPa s
Density at 23 °C		1.14 g/cm <sup>3</sup>

Table 1 Silicone properties from manufacturer product sheet [26].

#### **4.1.2 Filler (Aluminum Oxide):**

It is a white colored powdered with 95.5% pure Aluminum Oxide. It has high thermal conductivity (K) of 31 W/m.k and for its ability to interact with resins and enhance conductivity of polymer composite structures. In addition to these , there are also key properties it offers for composite fabrication such as availability in powder form with low agglomeration property that helps good dispersion in the matrix, resists strong acid and alkali attacks ,excellent size and shape capability, non corrosive, non-toxic property and low cost. Hence for our purpose Aluminum oxide has been selected as the filler material [40].

### **4.2 Lab Fabrication of particulate filled composite**

To develop particulate filled composite material by blending low viscosity Silicone Elastomer with Aluminum oxide by predefined proportion of sample preparation. And to measure the thermal conductivity value of the prepared samples and to identify the most suitable composition for the application of thermal bridge material that fills uneven surface of a building walls to facilitate thermal flux measurement.

#### **4.2.1 Matrix Preparation**

Before mixing Aluminum Oxide powder ( $Al_2O_3$ ) with silicone resin to make the composite product, blend of Silone resin with viscosity of  $1.14g/cm^3$  and the catalyst is prepared as a base matrix that cures in room temperature. The two types of elastosil M resins with their respective catalyst included in the package.



Figure 4 Low viscosity silicone resins and catalyst from Wacker chemical

To prepare the catalyzed mix, the resin had to be mixed with the recommended catalyst by the manufacturer, before filler material Al<sub>2</sub>O<sub>3</sub> is incorporated. Room temperature vulcanized (RTV) rubber is the result after mixture is cured with incorporation of Aluminum oxide powder resulting in composite sample.

The table summarizes the standard proportions of catalyst that should be added to get room temperature cured silicone pads.

	<b>Elastosil 4511</b>	<b>M</b>	<b>Elastosi 4601A</b>	<b>M</b>	<b>ElastosilM,B(catalyzed)</b>
catalyst T21(w%)	5		-		-
catalyst T51(w%)	5		-		-
catalyst T47(w%)	5		-		-
			9		1

Table 2 Standard Resin preparation according to manufacturer recommendation (26).



Figure 5 Addition vulcanization of RTV-2 silicone resin.

The amount of the base matrix silicone Elastosil and the respective catalysts to be used in order to get RTV (room temperature cured) silicone rubber pads or sheets molding. Before the curing process start it is recommended to use the catalyzed mix to blend with the filler particles as faster as possible, since the viscosity increases it can result in difficulty to incorporate the filler material and obtain uniformly dispersed in the matrix that affects the characteristics of the final composite.

#### **4.2.2. Filler ( $\text{Al}_2\text{O}_3$ ) incorporation**

1. The mass of the filler material alumina powder ( $\text{Al}_2\text{O}_3$ ) calculated according to the required weight fractions of the filler materials which are; ( 5,10,15,20,30,40) wt%. of the resin (silicone Elastomeric). Sample composition is prepared by measuring the weight fraction (% wt) of the filler  $\text{Al}_2\text{O}_3$  and mixing with the weight fraction of the silicone resin (% wt).

Density of Alumina ( $\text{Al}_2\text{O}_3$ ) is  $4\text{g/cm}^3$ , and conversion the mass to volume fraction enable us to know what amount of gram to mix to obtain the required volume fraction.



Figure 6 Aluminum oxide powders as thermal conductive filler.

2. The mass of the resin (silicone elastomer) is mixed according to the required volume of blend and the size of the composite material required. In addition to this the density of the composites could be determined from equation (6).

Filler (w %)	matrix (g)	$V_f(\%)$	$V_m(\%)$	Composite Density ( $\rho_c$ )
5	50	1.5	98.5	1.18
10	50	3.1	96.9	1.23
15	50	4.8	95.2	1.28
20	50	6.7	93.3	1.33
30	50	10.9	89.1	1.45
40	50	16	84	1.6

**Table 3** Sample preparation by varying the amount of filler percentages by weight.





Figure 7 Mixture of RTV-2 Silicone after through stirring.

3. Accelerator (catalyst) and the hardener are added as weight % according to the manufacturer recommendation given in the product sheet.
4. The filler and the catalyzed matrix are mixed for about 10 minute at room temperature continuously and slowly to avoid bubbling during mixing, and then  $Al_2O_3$  filler is added to the mixture with gentle mixing until uniformly distributed.
5. The mixture is poured from one corner into the mould (to avoid bubbles formation which causes air to hold) and the uniform pouring is continued until the mould is filled to the required level.
6. The mixture is left in the mould for (24) hrs at room temperature to solidify. Then the cast is placed inside a room temperature.



Figure 8 Mixture allowed curing at room temperature.

8. The specimens are de-molded accordingly and cut in to the standard dimensions for a test.
9. Thermal conductivity test is conducted on each sample using flux meter .After this test the thermal conductivity  $k$  value of each samples were determined.

### 4.3 Thermal conductivity analysis of Samples

Thermal conductivity of pre-formulated composite samples were analyzed based on both theoretical models and experimentally determined values. The theoretical values were calculated using Maxwell model and using equation (13), to determine the effective thermal conductivity ( $k_{ef}$ ) of randomly dispersed filler particles in the matrix. Additionally experimental values were obtained using measurements obtained by using heat flux meter found at Arcada.

The average thickness of the specimen is 1.5mm, and the U values are given from the device readings. In addition to this heat flux is calculated by the relation:

$$q' = \frac{q}{A} = k * \frac{\Delta T}{L}$$

Using equation 3 and substituting heat flux  $dq'$  values obtained from the device gives us the thermal conductivity of the samples.

$$k = \frac{dq' \cdot L}{dT}$$

The  $dq'$  values are given in the table below while; the thickness  $L$  for all the samples is 1.5mm and the area  $A$  is the heat transfer area that is equal to the area of the flux-meter and it is  $0.01m^2$ . Temperature change was recorded during the measurement on the outer surface of the composite and inside surface,  $T_{out}=21.6^{\circ}C$ ,  $T_{in}=21.3^{\circ}C$ , so  $dT=0.3^{\circ}C$ .

### U-value calculation

An experimental result based on measurements taken by heat flux meter was given by  $q$  values in **milliWatts (mW)**. First “Dry Runs” have been made to get mean U values of the wall, and then the actual test was made by placing the silicone between the wall and the meter. The ‘dry run’  $q$  value was found to be **50 mW**, is the flux from the wall surface on the area  $A=0.01m^2$ . The mean U-value of the wall is,

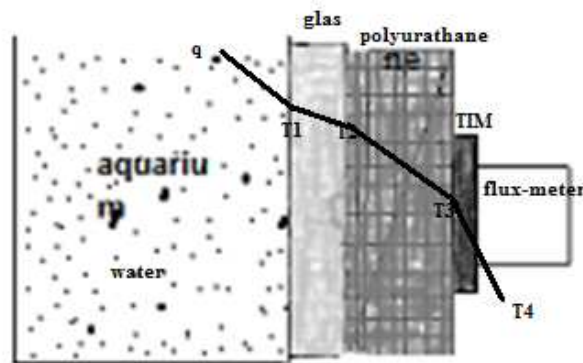
$$q = U_{\text{wall}} * A * \Delta T \quad \Delta T, \quad T_{\text{water}} = 7.8^{\circ}\text{C}$$

$$U_{\text{wall}} = \frac{q}{A * \Delta T} \quad T_{\text{out surface}} = 21.6^{\circ}\text{C}$$

$$U_{\text{wall}} = \frac{50mW}{0.01m^2 * (21.6 - 7.8)^{\circ}\text{C}}$$

$U_{\text{wall}} = 0.36 \text{ W/m}^2.k$ , the mean U-value of the wall without silicone TIM

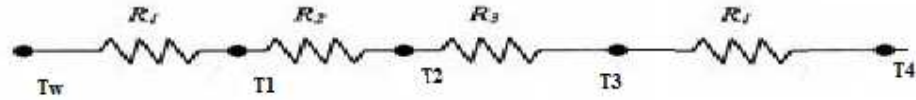
### U-value calculation from simulated aquarium wall layers using Silicone.



**Figure 9** Simulated wall layers for experimental measurement of heat and U values.

As the figure above shows, the wall is constructed from three layers, glass layer, polyurethane layer and the TIM layer, and it also shows the flux meter.

To calculate the overall U values from these wall layers, the corresponding thermal circuit is shown below.



Since,  $R_{tot} = 1/(U_{tot} * A)$

The overall thermal transmittance equation is given by:

$$\frac{1}{U} = \frac{1}{h_w} + \frac{L_{glass}}{k_{glass}} + \frac{L_{poly}}{k_{poly}} + \frac{L_{TIM}}{k_{TIM}}$$

However; since the effect of water in the tank has small effect on the system, it can be ignored and the above equation will be given as

$$\frac{1}{U} = + \frac{L_{glass}}{k_{glass}} + \frac{L_{poly}}{k_{poly}}$$

Known thermal property values are;

$T_w$  = inside water temperature = 7.5°C

$K_{polyurathane}$  = 0.03

$L_{polyurathane}$  = 35mm = 0.035m

$K_{glass}$  = 0.96 W/m-k

$L_{glass}$  = 5mm = 0.005m

Illustrating as example below for sample 1,

$$\frac{1}{U} = \frac{L_{glass}}{k_{glass}} + \frac{L_{poly}}{k_{poly}}$$

$$\frac{1}{U_{wall}} = \frac{0.005}{0.96} + \frac{0.035}{0.03}$$

Therefore,  $U_{wall}=0.85W/m^2.k$

The difference between the values from the flux-meter and from calculation is found to be 0.4.

The experimental U-value for each sample is calculated accordingly and the result can be referred from table 4.

Samples	(% wt) Filler	Theoretical $k_{ef}$ Values (W/m-k)	Theoretical U-value of samples (W/m <sup>2</sup> -k)
1	Resin+5% Al <sub>2</sub> O <sub>3</sub>	0.21	140
2	Resin+10% Al <sub>2</sub> O <sub>3</sub>	0.22	146
3	Resin+15% Al <sub>2</sub> O <sub>3</sub>	0.23	153.3
4	Resin+20% Al <sub>2</sub> O <sub>3</sub>	0.24	160
5	Resin+30% Al <sub>2</sub> O <sub>3</sub>	0.27	180
6	Resin+40% Al <sub>2</sub> O <sub>3</sub>	0.3	200

**Table 4** Theoretical and experimental results of k and u value based on the composite samples.

### Specific Heat determination of the composite samples

Specific heat capacity of the samples were also determined by using equation 8

$dq = Cm\Delta T$ , the mass (m) of the each sample were found by weighing (g). Example on the first sample is illustrated as follow,

$$dq = 41 \text{ mW}$$

$$\text{Mass (m)} = 52,5\text{g} , \Delta T = 0.3^\circ\text{C},$$

Substituting values in  $dq = Cm\Delta T$

$$C_{\text{sample 1}} = dq/m\Delta T = 0.041\text{Watts}/(0.0525\text{Kg} * 0.3^\circ\text{C})$$

$$= 2.62 \text{ J/kg-}^\circ\text{C}$$

Filler (w %)	Matrix (g)	Mass (g) of composite	Experimental dq values (mW)	Specific heat capaci- ty (C) in (J/kg-°C)
5	50	52.5	41	2.6
10	50	55	38	2.3
15	50	57.5	31	1.8
20	50	60	28	1.55
30	50	65	24	1.23
40	50	70	21	1.0

**Table 5** Specific heat Capacity of the composite samples.

#### **4.4 Characterization of the composite Materials**

After first, trial of matrix preparation based manufacture's instruction and incorporation of filler, the resulting cured silicone composite did not fulfill the parameters required for the desired purpose. For instance, it was found tough that has lower flexibility that could not fill the roughness.

The main important characteristics of this material development is to obtain a material as soft as possible to be able to fill a air gaps on the surface of a building walls, so that it must be as conformable as possible, that means, flexible and soft enough, in addition the material should be able to stick to the wall that can be removable at any time without leaving a stain or spot on the wall and user friendly without causing any hazard.

Having these important properties in mind, silicone elastomeric was chosen as a matrix material. Adding  $\text{Al}_2\text{O}_3$  fine powdered into the catalyzed silicone matrix was one of the important steps in fabrication process. After addition, mixing thoroughly enables to obtain uniformly and randomly distributed filler particles in the matrix that prevents cluster formation in one place resulting in poor uniform density of material. Mixing also helps good surface texture and structural visibility. Hence, during mixing the two phase materials integrate so that thermal conduction is also be enhanced to a greater extent.



## 5 RESULT AND DISCUSSIONS

Having done the experiments in Arcada chemistry lab, polymer composite sample could be formulated based on the application requirement for thermal bridging purpose to be used as an interface material between rough surface of a building and thermal flux measuring meter that was designed earlier in the School to measure the energy dissipation from a building walls. The base matrix is prepared according to the above procedure by blending the silicone resin with the catalyst. And the preceding step was to mix the amount of Aluminum oxide powder needed by weight in to the catalyzed resin to form good dispersion and distribution by mixing.

After several trials and varying the amount of catalyst needed to cure the resin, it was found that soft, flexible and elastic polymer composite material samples could be formulated. At first higher amount of catalyst added, resulting in tough and harder sample after curing process is undergone. This property is not desirable for the required purpose, while step by step addition of catalyst with varying amount could result in desirable composite product. The amount of catalyst added was less than half of the recommended by the manufacturer 5%.

Experimental Thermal conductivity measurements using the flux-meter revealed that, increasing the amount of filler decreases the conductivity in a very significant manner; the result also shows contradictive figures with the theoretical value, this is because as the amount of filler percentage increases the toughness also increases resulting in incomplete conformability into the wall leaving air gaps, secondly as the filler increases there is also a tendency to form a multiple layer of particles in the matrix that results the heat transfer rate slow. However; this property is important for composite used for other applications where a toughness and hardness property of a material is significant and the theoretical value is universal for such composite materials.

The Aluminum oxide particulate filled in silicone composite shows good enough to fill the roughness of a wall, self tacky on the wall to mount the measuring device. It was found that higher Elasticity, flexibility and good surface texture were observed with lower filler loading. It was seen that increasing the amount of filler fraction decreases the softness and increases the rigidity that also decreases thermal conductivity.

## 6 CONCLUSION

Having frequent trials of sample preparation by varying the amount of catalyst (hardener) to be added, it was found softer, flexible, user friendly material that fills gaps on the rough surfaces to facilitate energy transfer in the form of heat through itself by avoiding air as thermal insulator.

Accordinging specific heat capacity ( $C$ ) results shown in table 4, the lower value shows that the property that is indicative of material's ability to absorb heat from the external surrounding wall with small energy and temperature difference.

Significant change in thermal conductivity value was not observed. This could be caused by addition of small amount of filler volume fraction in the matrix resin. In other cases, increasing the amount of filler volume fraction results to a material with undesirable characteristics such as hardness and toughness also increases. The better the material complies with both surfaces, the less air interstitials there are. However; even though small thermal conductivity change is observed in the test result, thermal contact resistances are mainly caused by incomplete fitting and air interstitials affecting the border areas with  $k_{Air} = 0,0027 \text{ W/m-K}$ , that acts as thermal insulator, but the composite conductivity values are much higher than the air to use as thermal interface. The better the material complies with both surfaces, the less air interstitials there are.

The chosen components the resin and conductive filler could be blended for the composite material development and achieved required characteristics such as thermal conductivity and stability, good filler-matrix integration, flexibility, surface conformability and tackiness as well as user friendly (non-toxic).

The samples were tested and used as interface material between surfaces and flux meter to fit the air gaps created by the rough surfaces to avoid thermal insulation, and positive result was obtained.

## 7 REFERENCES

1. Ye.P. Mamunya et al. I European polymer Journal 38, Electrical and thermal conductivity of polymers filled with metal powders. (2002) 1887-1897.
2. C.Luo, B.Moghtaderi, S.Hands, A. Energy and Buildings 43(2011) 379-385.
3. J.P. Gwinn, R.L. Webb, Microelectronics Journal 34, Performance and testing of thermal interface materials (2003) 215-222
4. Thermal interface materials, <http://www.electronics-cooling.com/1996/09/thermal-interface-materials-2/>, Accessed 10.02.2012.
- [5]. J.G.Poloniecki et al. Steady state analysis of Zero-balance heat flux meter, Sensors and Actuators A 49 (1995) (29-35).
- [6]. Jean L.Leblanc, Filled polymers science and Industrial applications, CRC press, Taylor and Francis group (2010).
- [7]. Peter Jerschow, Silicone Elastomer, wacker-chemie GmbH,  
<http://www.scribd.com/doc/76869343/Silicon-Elastometers>
- [8]. Robert H.Perry, Don green, Perry's Chemical Engineers Handbook, 6<sup>th</sup>ed, McGraw-Hill International Editions.
- [9]. Fran P. Incropera /Dewitt / et al, Fundamentals of Heat and mass transfer, 6<sup>th</sup>ed, John Wiley & Sons. Inc
- [10]. Francois Cardarelli, Materials handbook, a concise desktop reference ,2<sup>nd</sup> Ed,(2008).
- [11]. R.N.Rothon, Particulate filled polymer composite, Rapra Technology Limited, 2<sup>nd</sup> ed.
- [12]. Kirk-Othmer, Encyclopedia of Chemical Technology, Vol 11, John Wiley & Sons, (pdf)

- [13]. D.W Van Kreven, K.te Nijenhuis, Properties of polymers, Their correlation with chemical structure; their numerical estimation and prediction from Additive group contributions.
- [14]. Hala TIM partners, available,  
[http://www.hala-tec.com/mod\\_use/dokumente/Basics\\_TM\\_engl.pdf](http://www.hala-tec.com/mod_use/dokumente/Basics_TM_engl.pdf), accessed 14.03.2012.
- [15]. Bergquist, [www.bergquist.com](http://www.bergquist.com) , accessed 07.02.2012
- [16]. Combinatorial methods for polymer science, Conductive polymer composites, Vol.5 (pdf)
- [17]. [www.fujipoly.com](http://www.fujipoly.com)
- [18]. J.C. Maxwell, A treatise on electricity and magnetism, 3<sup>rd</sup> ed. Dover, New York (1954)
- [19]. M.Yovanovich, E.E Marotta., Thermal spreading and Contact resistances, available, [ftp://ftp.energia.bme.hu/pub/hoatadas\\_ket\\_fejezete/BEJAN,%20A.%20\(2003\).%20Heat%20Transfer%20Handbook/725\\_04.PDF](ftp://ftp.energia.bme.hu/pub/hoatadas_ket_fejezete/BEJAN,%20A.%20(2003).%20Heat%20Transfer%20Handbook/725_04.PDF) , (Accessed, 09.05,2012)
- [20]. S.W. Gregory, K.D. Freudenberg, P. Bhimaraj and L. S Schadler, A study on the friction and wear behavior of PTFE filled with alumina nanoparticles, J. Wear, 254 (2003) 573–580.
- [21]. K. Jung-il, P.H. Kang and Y.C. Nho, Positive temperature coefficient behavior of polymer composites having a high melting temperature, J. Appl. Poly Sci., 92 (2004) 394–401.
- [22]. S. Nikkeshi, M. Kudo and T. Masuko, Dynamic viscoelastic properties and thermal properties of powder-epoxy resin composites, J. Appl. Poly. Sci., 69 (1998) 2593-8.
- [23]. K. Zhu and S. Schmauder, Prediction of the failure properties of short fiber reinforced composites with metal and polymer matrix, J. Comput. Mater. Sci., 28 (2003) 743–8.
- [24]. M. Rusu, N. Sofian and D. Rusu, Mechanical and thermal properties of zinc powder filled high density polyethylene composites, J. Polymer Testing, 20 (2001) 409–17.
- [25]. L.A. Utracki, polymer blends Handbook, Kluwer, Volume 1 (2002)

[26].Wacker silicones,

<http://www.wacker.com/cms/en/products-markets/products/product.jsp?product=9125> ,

Accessed 09.05.2012..

[27]. S.T. Peters, Handbook of composites, 2<sup>nd</sup> ed., Chapman and Hall, London, 1998, pp. 242–243.

[29]. D. Kumlutas, I.H. Tavman, and M.T. Coban, Thermal Conductivity of Particle Filled Polyethylene Composite Materials, J. Composites Sci. and Tech., 63(1) (2003) 113–117.

[30]. I.H. Tavman, Thermal and Mechanical Properties of Aluminum Powder filled High-density Polyethylene Composites, Journal of App. Poly. Sci., 62(12) (1996) 2161–2167.

[31]. N.M. Sofian, M. Rusu, R. Neagu and E. Neagu, Metal Powder-filled Polyethylene Composites. V. Thermal Properties, J. of Thermoplastic Composite Materials, 14(1) (2001) 20–33.

[32]. Agarwal B D, Broutman L J. Analysis and performance of fiber composites: Second Edition. John Wiley and Sons, Inc.; 1990.

[33]. I. Tavman, Thermal Anisotropy of Polymers as a Function of their Molecular Orientation, Experimental Heat Transfer, Fluid Mechanics, and Thermodynamics, Elsevier (1991) 1562–1568.

[35]. N.M. Sofian, M. Rusu, R. Neagu and E. Neagu, Metal Powder-filled Polyethylene Composites. V. Thermal Properties, J. Thermoplastic Composite Materials, 14(1) (2001) 20–33.

[36]. Y.P. Mamunya, V.V. Davydenko, P. Pissis and E.V. Lebedev, Electrical and Thermal Conductivity of Polymers Filled with Metal Powders, J. European Polymer, 38(9) (2002) 1887–1897.

[37]. B. Weidenfeller, M. Hofer and F.R. Schilling, Thermal Conductivity, Thermal Diffusivity, and Specific Heat Capacity of Particle Filled Polypropylene, J. Composites Part A: Applied Science and Manufacturing, 35(4) (2004) 423–429

[38]. H.S. Tekce, D. Kumlutas, and I.H. Tavman, Determination of the Thermal Properties of Polyamide-6 (Nylon-6)/Copper Composite by Hot Disk Method, In: Proceedings of the 10th Denizli Material Symposium, Denizli, (2004) 296–304.

[39]. Hyungu Im • Jooheon Kim, Enhancement of the thermal conductivity of aluminum oxide–epoxy terminated poly(dimethyl siloxane) with a metal oxide containing polysiloxane, J Mater Sci (2011) 46:6571–6580.

[40]. Aluminum Oxide, <http://accuratus.com/alumox.html>, Accessed 12.04.2012

41. S.L.Kakani, Amit Kakani, Material Science book, New Age International Ltd (P), 2004.