# The effect of thermal aging on the properties of recycled copper filled epoxy/unsaturated polyester composites

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**ABSTRACT**

A blend system consist of two types of thermoset polymer, epoxy and unsaturated polyester at ratio of 80/20 weight percentage (wt. %) filled with 0, 20 and 40 volume percentage (vol. %) of recycled copper powder as conductive filler was prepared and undergone thermal aging process for 40 days. The changes on volumetric dimension of the blend composites were studied. The aged blend composites were undergone characterization and testing which include flexural, hardness, volumetric shrinkage and electrical conductivity properties. After thermal aging, the flexural strength of the blend composites increased started from 20 vol.%of filler loading. Besides that, the hardness properties and electrical conductivity were also improved after thermal aging.

Keywords: Polymer blend, Recycled copper, Thermal aging, Epoxy, Unsaturated polyester

## 1.0 INTRODUCTION

Conductive polymer composites are fabricated from the mixing of insulating electrical properties of polymer matrix and electrical conductive properties from conductive fillers, such as carbon black or metal powder[1-6]. When a polymer is exposed to a relatively small amount of heat over a long period of time, the cumulative effects can be equalize to high temperature degradation. The polymer is said to reach its thermal stability limit and begin to degrade, it is also known as thermal degradation or thermal aging [7-10]. Chemically and physically, the process of aging is irreversible with time.

Several studies of thermal aging effect on both epoxy[11-13] and unsaturated polyester [14-16] have been reported, Birger et al. [17] has investigated on the effect of thermal aging on epoxy and found that thermal aging at 170°C causes a reduction in the flexural properties but the study on epoxy/unsaturated polyester blend composites are limited. The effect of thermal aging on the mechanical, thermal and electrical properties between epoxy/unsaturated polyester and recycled copper was determined. In this study, recycled copper filled epoxy/unsaturated polyester composites were prepared and thermal aged for a time period of 40 days. During the thermal aging process, the volumetric shrinkage of the blend composites were studied as well as the mechanical and electrical properties of the blend composites after thermal aging.

## 2.0 EXPERIMENTAL

### 2.1 Sample preparation

In this research, the thermoset blend system which were consisted of epoxy (clear epoxy resin DER331) and unsaturated polyester, both were supplied by Hasrat Bestari Sdn. Bhd. The epoxy/unsaturated polyester weight percentage ratio was fixed at 80/20. Epoxy and unsaturated polyester were cured with clear epoxy hardener at 60 phr and methyl ethyl ketone peroxides (MEKP) at 1 wt.% respectively, both materials were supplied by Hasrat Bestari Sdn. Bhd. Recycled copper powder was collected from mill machine as the waste. Epoxy, unsaturated polyester and recycle copper filler were mixed using mechanical stirrer at 1200 rpm for 3 minutes to achieve homogeneous mixing. After that, epoxy hardener clear was added and stirred for another 1 minute before MEKP was added. The amount of recycled copper powder added ranged from 20 and 40 volume percentage(vol.%). The mixture was then cast inside a polypropylene mold and cured under 100°C for 1 hour using a conventional air oven.

## 2.2 Thermal aging

Samples for each different blending ratio and filler volume fraction were prepared and cut into bar shape and thermal aged in the furnace at 150°C for 40 days, which approximately equal to 960 hours. The samples were aged in the heating condition of 150°C in order to accelerate thermal aging and attain a boundary limit, which is the beginning of thermal degradation [18].

## 2.3 Characterization

### 2.3.1 Flexural properties measurement

The flexural test was done according to ASTM D790. The 3 point flexural test was carried out by using Inston 5569 Universal Testing Machine (UTM), with cross-head speed of 2.38mm/min. The flexural strength and flexural modulus were determined based on the Equation (1) and (2), under a load in a three point bending setup.

$$\text{Flexural stress, } \sigma_f = \frac{3PL}{2bd^2} \qquad \text{- Equation (1)}$$

$$\text{Flexural modulus, } E_f = \frac{L^3 m}{4bd^3} \qquad \text{- Equation (2)}$$

where;
$\sigma_f$ = flexural stress, MPa
$E_f$ = modulus of elasticity in bending, Mpa
P = load at a given point on the load-deflection curve, N
L = support span, mm
b = width of beam tested, mm
d = depth of beam tested, mm
m = slope of the tangent to the initial straight-line portion of the load-deflective curve, (N/mm) of deflection

### 2.3.2 Hardness measurement

The hardness properties of the recycled copper filled epoxy/unsaturated polyester composite was tested using Vickers hardness tester FV-700e, with load of 1kg (9.8N) for 15 seconds.

### 2.3.3 Volumetric shrinkage measurement

The volumetric shrinkage for each sample was measured every 10 days, approximately 240 hours for a total of 40 days. The volume of each samples were measured every 10 days under condition of sample’s temperature at 24±2°C.

### 2.3.4 Electrical properties measurement

The electrical resistivity and conductivity properties of the samples were measured by using Kyoritsu Digital Multimeter 1009. Each samples were coated with a layer of conductive silver paste and current was pass through a resistor before to the sample and sample's resistivity was measured between the samples. Electrical resistivity is the reciprocal of electrical conductivity, and the electrical conductivity of each different filler loading sample is calculated by dividing the sample thickness by the product of each sample resistance and its cross section area. The electrical conductivity of each specimen was then calculated using Equation (3).

$$\sigma = \frac{h}{AR}$$ - Equation (3)

where,

h = sample thickness
A = cross-sectional area
R = resistance

## 3.0 RESULTS & DISCUSSION

### 3.1 Flexural properties

In Figure 1 and Figure 2, it shows the flexural strength and modulus of the recycled copper filled epoxy/unsaturated polyester before and after thermal aging. From the figures, the unfilled epoxy/unsaturated polyester composite has undergone minor reduction in flexural strength after thermal aging. This observation may due to the high weight percent of epoxy in the blend ratio, which epoxy has undergone thermal degradation and reduction in strength due to some chain scission and surface oxidation which lead to weaken in mechanical properties [19].

It is interesting to note that after thermal aging, the flexural strength improved after thermal aging. With further increase of the filler loading up to 40 vol.%, the flexural strength is reduced after thermal aging. These phenomena can be explained by the thermally induced post-cure of thermosets during aging process, which the chain mobility was reduced [19] at 20 vol.% filler loading, and hence, the flexural strength increased as compared to without thermal aging. However, due to the significant different in thermal expansion coefficients between the recycled copper and epoxy/unsaturated polyester composites, this has resulted the heat build-up of thermal stresses between the fillers and polymer composites [17]. The induced thermal stresses may result in crack formation between matrix-matrix phases, matrix-filler phases and filler-filler phases, which will lead to the degradation of mechanical properties of the blend composites. It is believed that, more thermal stress was generated with increasing of recycled copper filler content and hence reduced the flexural strength at 40 vol.% as compared to un-aged sample at the same filler content.

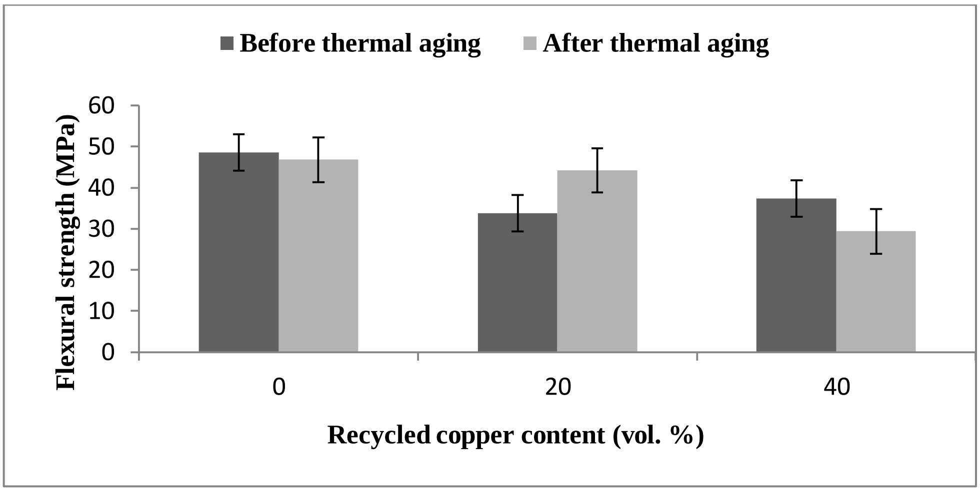


Figure 1: Flexural strength of recycled copper filled epoxy/unsaturated polyester composites before and after thermal aging

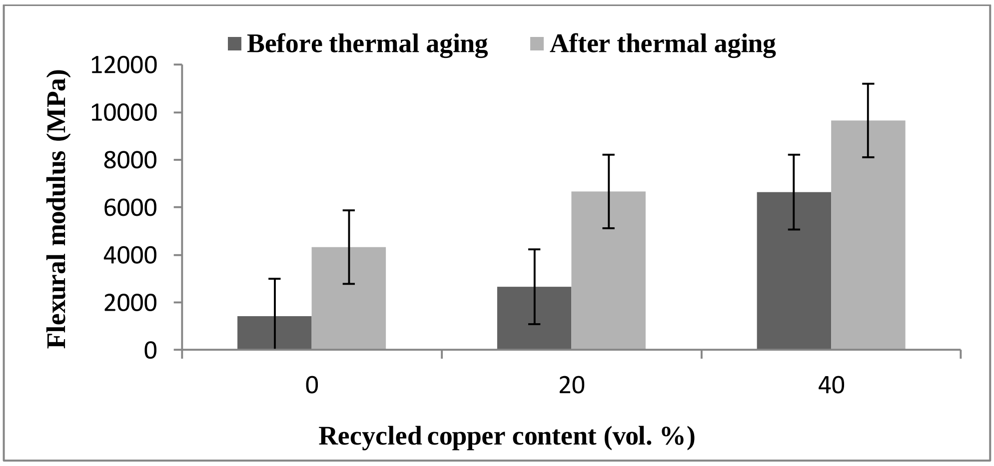


Figure 2: Flexural modulus of recycled copper filled epoxy/unsaturated polyester composites before and after thermal aging

For flexural modulus of the recycled copper filled epoxy/unsaturated polyester composites in Figure 1 and Figure 2, the results show an increment for each of the different filler loading after thermal aging. This may due to the thermal aging effect, which the molecule chains in every blending ratio was packed up and tighten closely, which increase the flexural modulus of the blend composites [19].

3.2 Hardness

Figure 3 shows the Vickers hardness of the recycled copper filled epoxy/unsaturated polyester composites, comparing before and after thermal aging. From the result, the Vickers hardness of the blend composites improved after 40 days of thermal aging. Due to thermally induced post-cure [19], which lead to post crystallization and shrinkage mechanism to occur, hence, the polymer chains are realigned to form crystallization zone. Post-cure process will increase the crosslink density of the blend composites and the chain mobility of the blend composites is reduced which resulted in improvement of resistivity of the blend composite's surface deformation or hardness test.

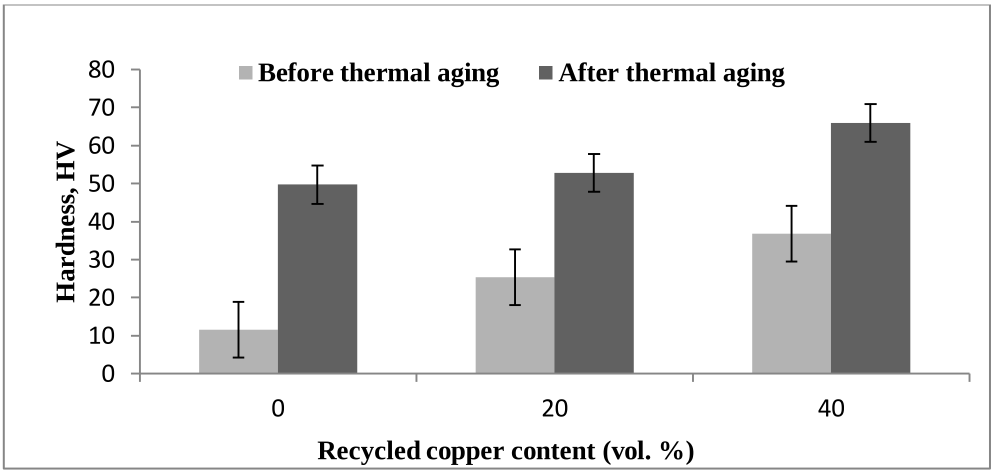


Figure 3: Vickers hardness of recycled copper filled epoxy/unsaturated polyester composites before and after thermal aging

3.3 Volumetric shrinkage

From previous study [20], it was shown that the magnitude of the volumetric shrinkage was greatly influenced by its filler volume fraction, the composition and the degree of conversion of the resin matrix. In Figure 4, it shows the volumetric shrinkage percentage of recycled copper filled epoxy/unsaturated polyester composites after thermal aging. From the figure, the unfilled epoxy/unsaturated polyester composite showed a higher volumetric shrinkage percentage compared to 20 and 40 vol.% recycled copper filled epoxy/unsaturated polyester composites. The introduced of recycled copper fillers into the epoxy/unsaturated composite resulted in reduction of free volume in the composite, where the distance between the molecules was greatly reduced. When the composite was undergone post polymerization or post crosslinking process, minor volumetric shrinkage was observed since the free volume was already occupied by the fillers. From the graph, it also indicated higher filler loading resulted in lower volumetric shrinkage, since more free volume in the composite was filled up when filler loading increased.

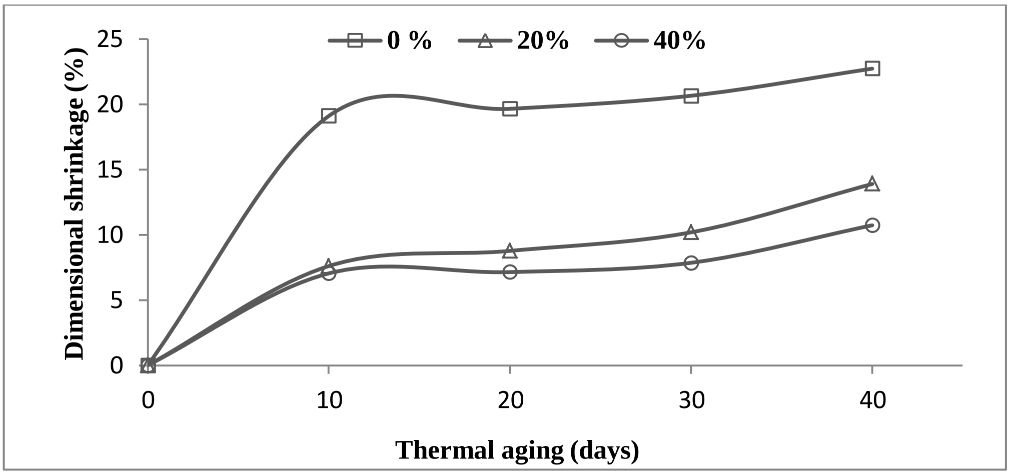


Figure 4: Volumetric shrinkage percentage of recycled copper filled epoxy/unsaturated polyester composites after thermal aging

3.4 Electrical properties

Figure 5 and Figure 6 show the electrical resistivity and conductivity of the recycled copper filled epoxy/unsaturated polyester at various filler loading after thermal aging. From Figure 5, a reduction in the electrical resistivity was observed after thermal aging. As for Figure 6, it shows an increment in the electrical conductivity of the blend composites after thermal aging.

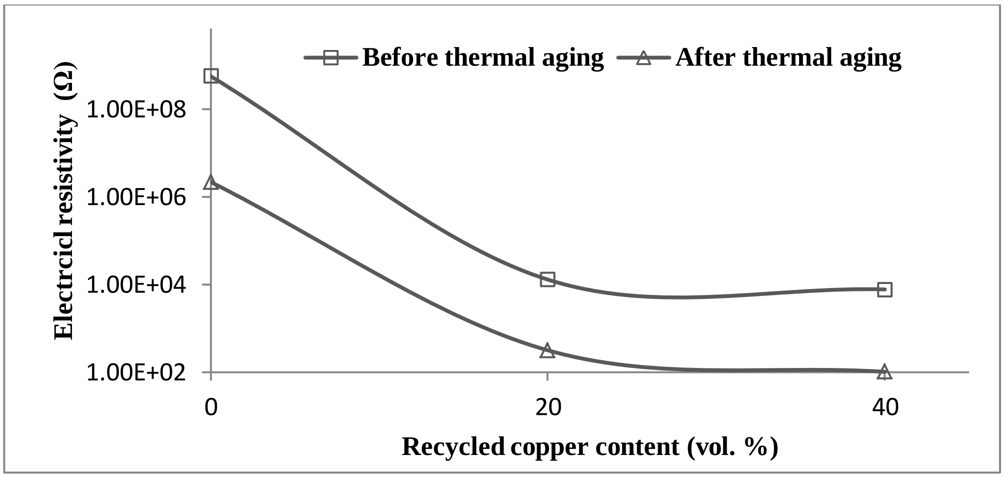


Figure 5: Electrical resistivity of the recycled copper filler epoxy/unsaturated polyester at various filler volume loading after thermal aging

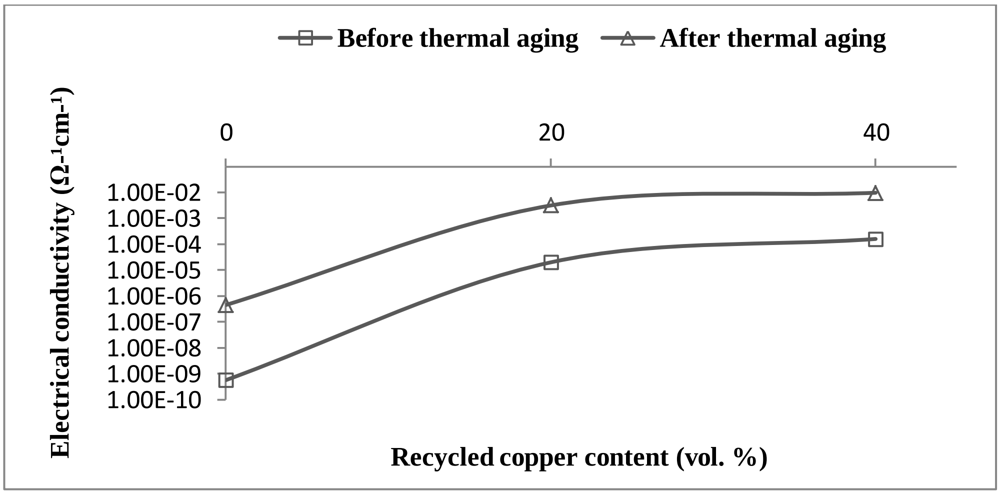


Figure 6: Electrical conductivity of the recycled copper filled epoxy/unsaturated polyester at various filler volume loading after thermal aging

The increment of electrical conductivity is believed to be due to the shrinkage of the sample after thermal aging. As the shrinkage increase, the distance between the recycled copper fillers were getting closer, hence, a decrease in electrical resistivity of recycled copper filled epoxy/unsaturated polyester composites was found. The same founding was reported by Li & Wong [21] in the polymer composite system.

## 4.0 CONCLUSION

In this research, the effect of thermal aging toward recycled copper filled epoxy/unsaturated polyester composites was studied. From the result, it observed that thermal aging can degrade the mechanical properties of the blend composite, an irreversible process where the blend composites shrink volumetrically at the same time improve the electrical properties of the blend composites.

## ACKNOWLEDGEMENT

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