CHAPTER IV
THE THERMAL CONDUCTIVITY OF METAL OXIDE-FILLED EPOXY COMPOSITE

4.1 Abstract

In this study, the effects of types and particle size of metal oxides on the thermal conductivity of the metal oxide-epoxy composite were reported. Scanning electron microscopy (SEM) was applied to analyze the fractured surface of metal oxide-filled epoxy composite. The results indicated that dispersion of nano-size particles was better than micro-size particles. The thermal conductivity of composite filled with nano-size particles was greater due to the improvement in the particle dispersion leading to more effective conductive network formation.

Keywords: Metal oxide-filled epoxy composite; Zinc Oxide; Copper Oxide; Thermal conductivity; Nanoparticle; Microparticle

4.2 Introduction

Typical thermal conductivity values in W/mK for some common materials are 0.2-0.3 for polymers, 234 for aluminum, 400 for copper, and 600 for graphite. Polymeric materials are typically limited in applications due to their inherent low thermal conductivity, low thermal stability, high electrical resistivity, and ductile mechanical properties. However, polymers may be attractive because they have high strength to weight ratio, are inexpensive, and are easy to process. One approach to improving the thermal conductivity of a polymer is through the addition of a conductive filler material, such as carbon, metal, and ceramics. In a polymer containing conductive fillers, heat is transferred by two mechanisms, lattice vibrations and electron movement. The addition of an additive into polymers may drastically enhance their properties and consequently their multifunctionality. This composite has paved the way for advanced technologies, such as electrochemical displays, sensors, catalysis, redox capacitors (Malianuskas, 2001).

Neshpor *et al.*, 1968 investigated the effect of the atomic number of metal on the thermal conductivity of those compounds. Their results show that the lattice thermal conductivity for many compounds increases with increase in their atomic number.

Sim *et al.*, 2005 found that the thermal conductivity of ZnO-filled composite was greater than that of Al_2O_3-filled composite. They suggested that the finer ZnO particles were more easily diffused in the polymer network compared to Al_2O_3 fillers.

In 2009, Lee and Dai studied the improvement in thermal conductivity of ethyl vinyl alcohol (EVA) composites filled with untreated and treated ZnO. They found that the thermal conductivity of EVA composites filled with untreated ZnO was greater than that of unfilled composite because the filler particles dispersed well in the matrix. However, the thermal conductivity of ZnO-filled EVA composite could be enhanced further by surface treatment due to the improvement in the interfacial adhesion between EVA matrix and ZnO particle.

The objective of this study is to investigate particle dispersion and the thermal conductivity of metal oxide-filled epoxy composite using two different metal oxides, ZnO and CuO, with.different particle sizes.

4.3 Experimental

4.3.1 Materials

The epoxy resin, bisphenol-A-(epichlorohydrin) (EPOTEC YD128), and the curing agent, cycloaliphaticamine (TH7301), were obtained from Aditya Birla Chemicals (Thailand) Ltd. ZnO (nano-size and <1 micron) and CuO (nano-size and <5 micron) were supplied by Fluka.

4.3.2 Preparation of Composite

The metal oxide was first dried in an oven at 100 °C for 2 h prior to processing. The desired amount of filler was gradually added to 60 mL of epoxy resin contained in a 250 mL plastic beaker. The loading of metal oxide powder was varied from 0.5%, 1.0% to 2.0 vol%. The mixture was then stirred by a mechanical stirrer (A.L.C. International S.r.l. Class I) at the mixing speed of 80 rpm for 15 min. Next, 36 mL of curing agent was added into the mixture which was then stirred until homogeneous. The mixture was then poured into a stainless steel mold 70 x 90 x 3 mm^3 in dimensions. The open mold was then placed in a vacuum oven at 50 °C for 10 min to evacuate the entrapped air. The mold was then closed by a 26x26 cm^2 metal plate and placed in the compression molding machine under 15 tons of loading for curing at 80 °C for 1 h.

4.3.3 Thermal Conductivity Measurement

Thermal conductivity of the composites was measured using a Hot Disk thermal analyzer (Hot Disk AB, Uppsala. Sweden). A minimum of three individual measurements were performed for each specimen (20 x 20 x 3 mm^3) with the sensor (3 mm diameter) being placed between two similar slabs of material. The sensor supplied a heat-pulse of 0.03 W for 15-20 s to the sample and the associated change in temperature was recorded. The average value of three specimens per sample was reported.

4.3.4 The Particle Dispersion Characterization

To determine the particle dispersion of metal oxide in epoxy matrix, the metal oxide-filled epoxy specimen was placed in liquid nitrogen for one minute. It was then broken into small pieces and one of the pieces was placed on a stub and examined using JEOL scanning electron microscopy (SEM) model JSM-5200 (Japan).

4.4 Results and Discussion

4.4.1 Correlation between Thermal Conductivity and Filler Content

Figure 4.1 and 4.2 show that thermal conductivity of metal oxide-filled epoxy composite increased with increase in filler content for both micro-size and nano-size particles. The thermal conductivity of the epoxy resin was found at 0.216 W/mK., while the addition of either ZnO or CuO in the range up to 2 vol% led to an increase of about 20% to about 0.25 W/mK. This is due to the high conductivity of the metal oxide. The increase in the conductivity with increase in filler content was due to the increase in the ease in the forming of conductive network at high filler content. Lee *et al.* (2006) studied the thermal conductivity of composites containing a high filler content of AlN. They found that the packing fraction of filler increased resulting in an increase in thermal conductivity of the composites. Furthermore, the results in Figure 4.1 and 4.2 also show that improvement in the thermal conductivity of nano-size particle-filled composite was greater than that of micro-size particle. This may be due to the greater surface area of the nano-size particles leading greater contact area with the polymer matrix, hence greater heat transfer efficiency. Yu and Choi, 2003 also suggested micro-size particles could settle rapidly during processing leading to poor filler dispersion. Gowda *et a.l* (2010) studied the improvement in thermal conductivity of alumina in DI water and copper oxide in DI water. They found that the thermal conductivity increased with the increase in the volumetric fraction of nanofluids. However, by using sonication technique, they found that sonication gave the highest effective thermal conductivity enhancement because the effective thermal conductivity was influenced by the particle dispersion and the average size of the particles in the nanofluids. In addition, in comparing between thermal conductivity of nano-size ZnO-filled and CuO-filled epoxy composites, the results show that the thermal conductivity of ZnO-filled composite was greater than that of CuO-filled composite, as shown in Figure 4.3. This result reveals that the thermal conductivity of composite increased with increase in the atomic number of metal atom of the filler. Ghoneim *et al.* (1983) investigated the dependence of the thermal conductivity of glass on the type and concentration of transition metal oxides. They found that the thermal conductivity increased with the atomic weight of the transition element.

4.4.2 Particle Dispersion

The fractured surface of metal oxide-filled epoxy composite was investigated. Figure 4.4 and 4.5 show the SEM micrographs of ZnO-filled and CuO-filled epoxy composite, respectively. The results from SEM micrographs show that the agglomeration of micro-size particle was more than the nano-size particle and both ZnO and CuO fillers showed the same trend. Furthermore, these Figures also show that the dispersion nano-size particles was more homogeneous than the micro-size particles. Therefore, the forming of conductive network and particle dispersion in nano-size particles was more effective than in the case of micro-size particle resulting in an increase in thermal conductivity of composites. Yu *et al.* (2002) found that the thermal conductivity of polystyrene-aluminum nitride composite increased under a special dispersion state of filler in the composites by homogeneously surrounding polystyrene matrix particles with aluminum nitride particles. Moreover, from Figure 4.6, the SEM micrographs show that the dispersion of ZnO particles in epoxy matrix was better than the CuO particles resulting in greater improvement in the thermal conductivity of ZnO-filled epoxy.

4.5 Conclusions

In this study, we found that the thermal conductivity of epoxy material could be enhanced by using metal oxide as a filler with ZnO being more effective than CuO. In addition, the nano-size particles were found to be more effective in enhancing the thermal conductivity of the composite for both ZnO and CuO. This was due to the greater surface area and the more homogeneous dispersion state of the nano-size particles when compared to the micro-size particles.

4.6 References

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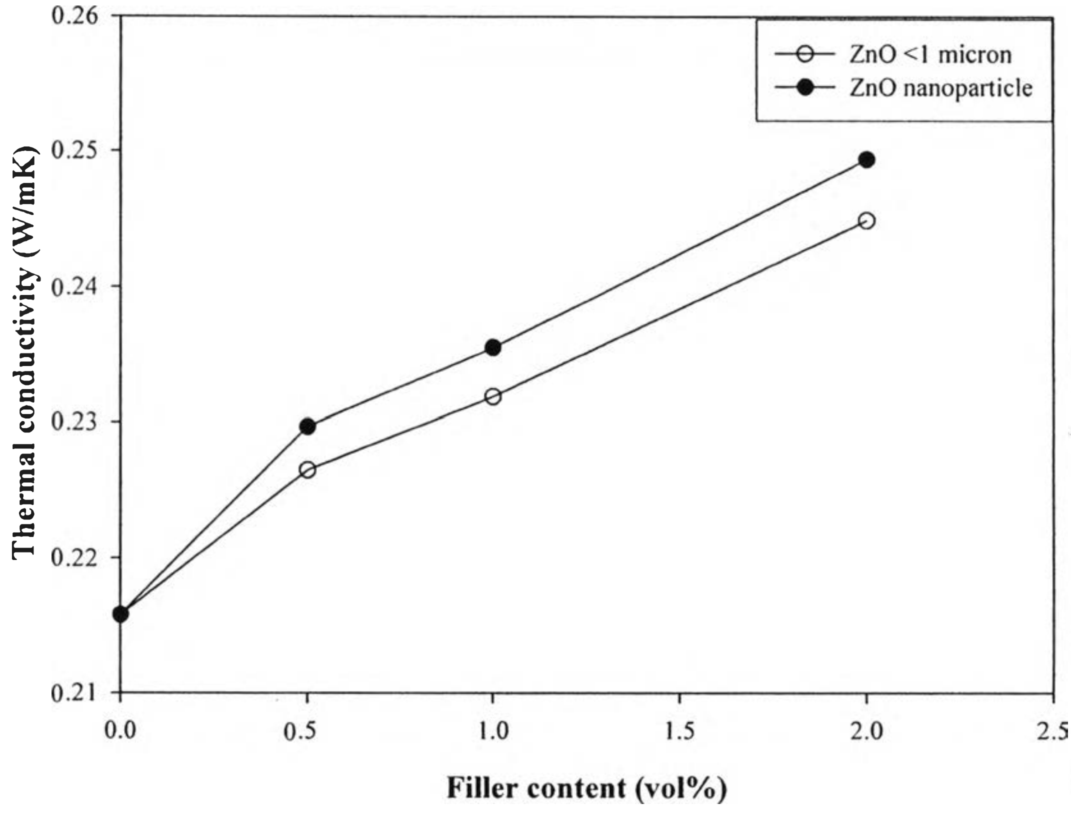

Figure 4.1 Thermal conductivity of ZnO-filled epoxy composite.

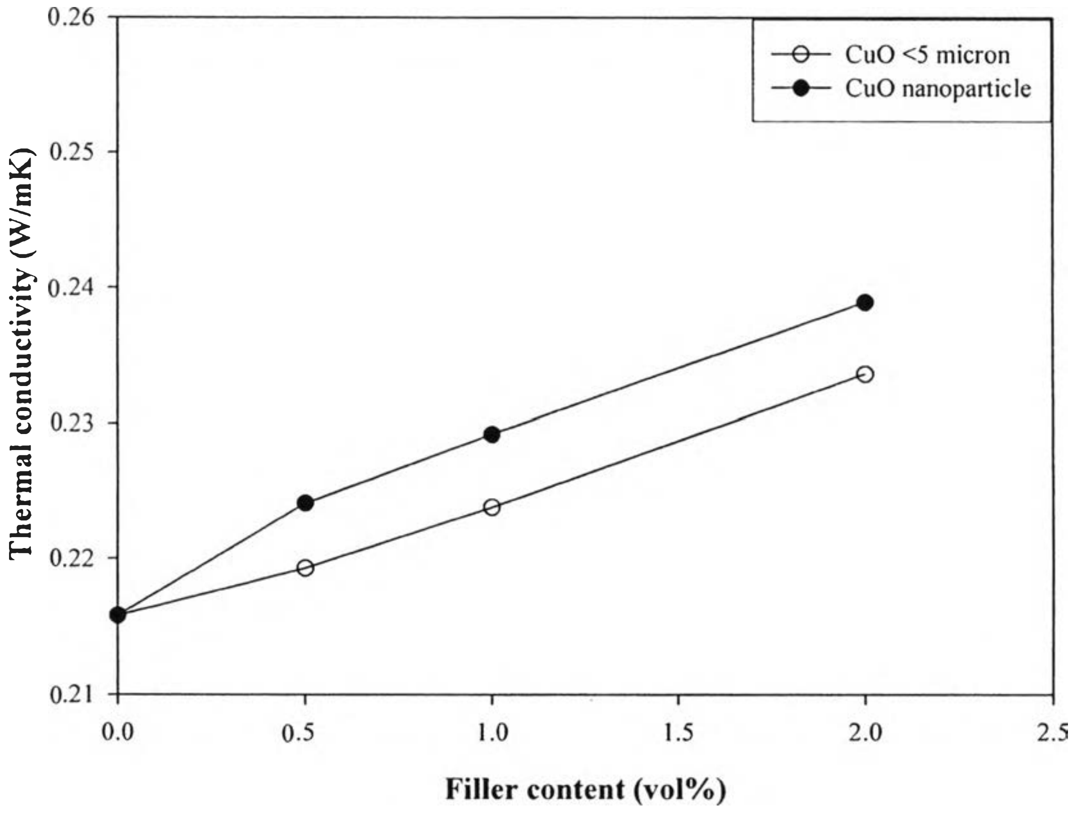

Figure 4.2 Thermal conductivity of CuO-filled epoxy composite.

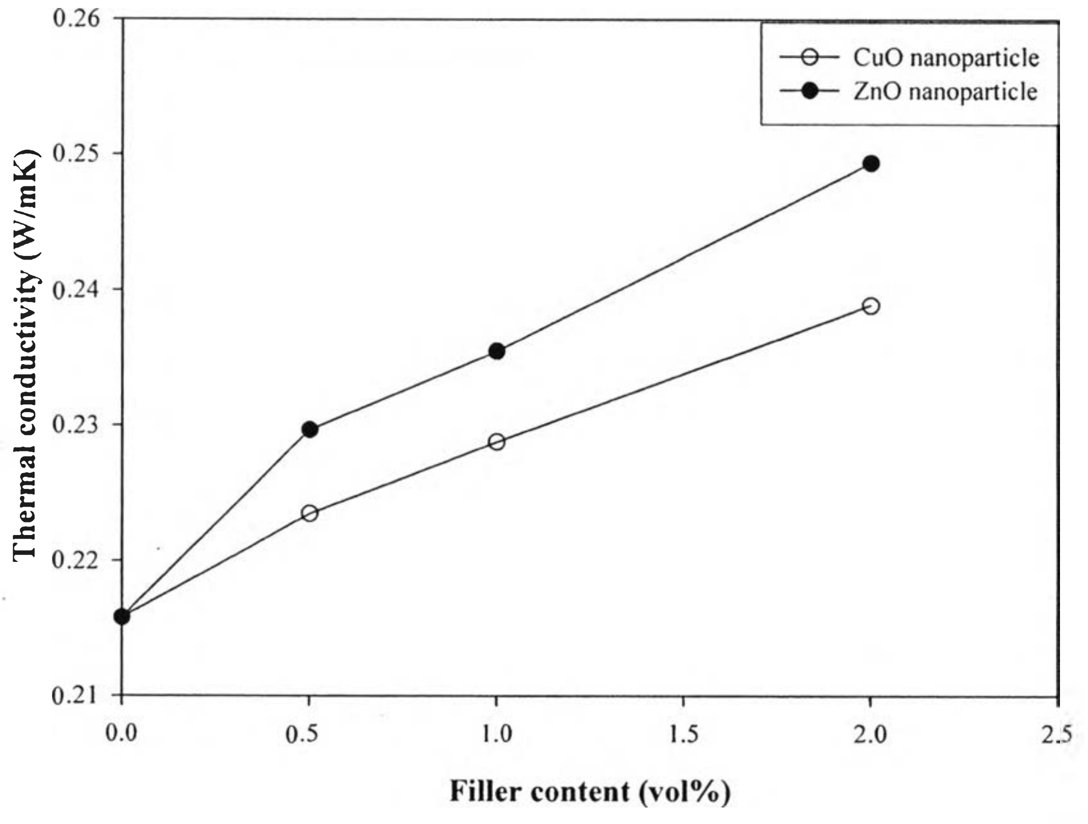

Figure 4.3 Comparison of the thermal conductivity of ZnO-filled and CuO-filled epoxy composite.

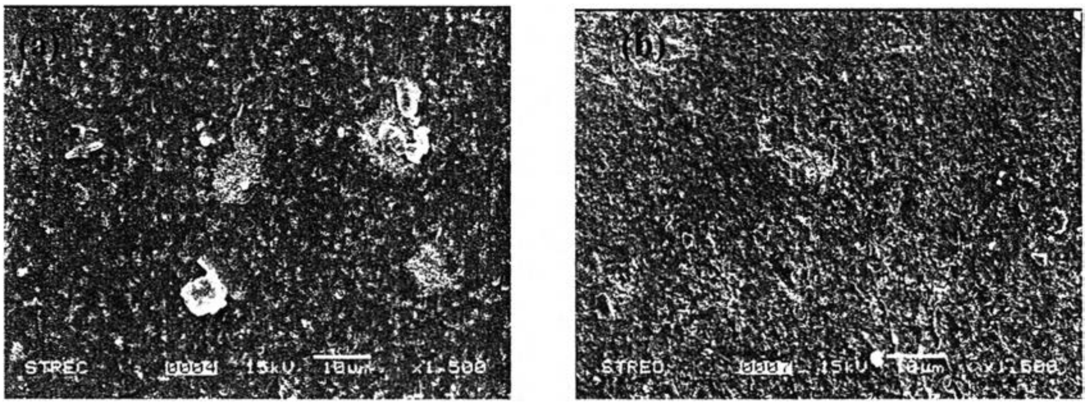

Figure 4.4 SEM micrographs of the fracture surface of (a) ZnO (< 1 micron) and (b) ZnO (nanoparticle) filled epoxy composite.

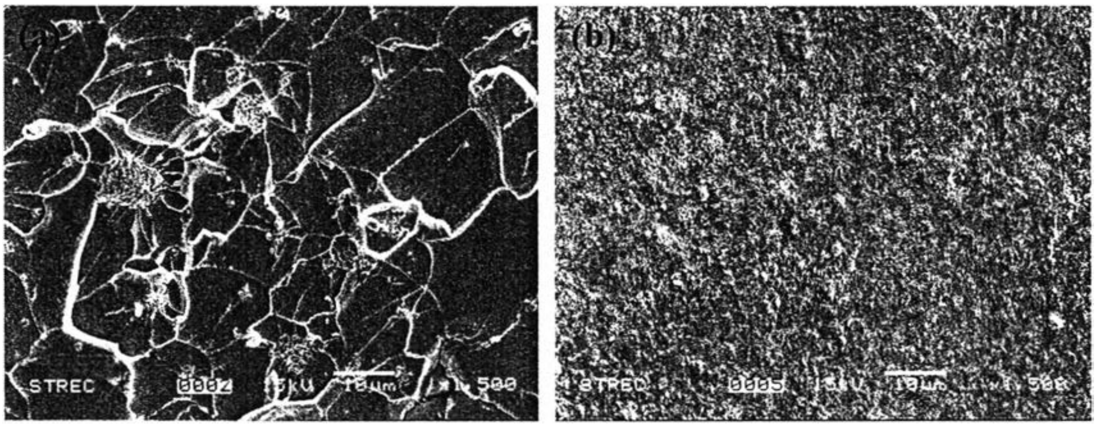

Figure 4.5 SEM micrographs of the fracture surface of (a) CuO (< 5 micron) and (b) CuO (nanoparticle) filled epoxy composite.

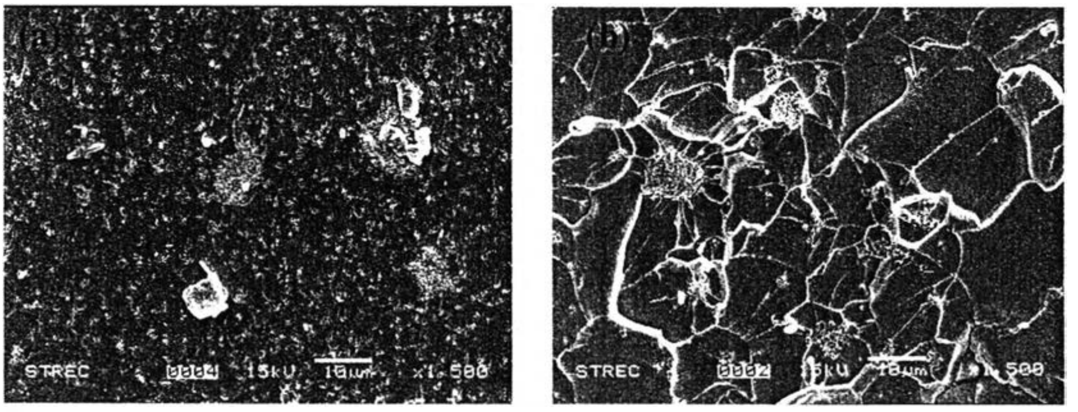

Figure 4.6 Comparison of particle dispersion between (a) ZnO-filled epoxy composite and (b) CuO-filled epoxy composite.