Microstructure, Microhardness and Thermal Properties of Aluminum with Multi-Walled Carbon Nanotubes Composites Prepared by Liquid State Processing

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Keywords: Microstructure, Microhardness, Thermal Conductiviy, Composites, Aluminum, Multi-Walled Carbon Nanotubes.

Abstract: Aluminum matrix composites reinforced with 0-10 wt.% copper-coated multiwalled carbon nanotubes (Cu/MWNTs) were produced by liquid state processing. The composites with < 10 wt.% Cu/MWNTs additions had higher thermal conductivity than the pure aluminum produced by the same liquid state processing. The Cu/MWNTs/Al composites exhibited the maximum thermal conductivity of 442.32 W/m/K at 8 wt.% Cu/MWNTs. The enhancement of thermal conductivity is supported by the measured microhardness. The Cu/MWNTs/Al composites exhibited the maximum microhardness 91,3 HV also at 10 wt.% Cu/MWNTs. The contribution of carbon nanotubes to thermal conductivity of the composites was demonstrated by theoretical analysis. The results show that copper-coated multiwalled carbon nanotubes (Cu/MWNTs) reinforced aluminum matrix composite is a potential material for high thermal conductivity applications.

1 INTRODUCTON

The increase in heat from electronic components is a major problem faced in electronic technology because of the miniaturization of these components (Ashby, et al., 2004). Heat sinks are used to dissipate the heat generated by these electronic components. A heat sink is described as an object that disperses or dissipates heat from another object. Usually heat sinks are widely used in computers and microelectronics as well as other applications (Reddy and Gupta, 2010).

Aluminum is the most commonly used material for heat sinks due to its light weight, lower cost, manufacturing capability, and infrastructure (Keller, 1998). The thermal conductivity of aluminum is about 220 W/mK (Dogruoz and Arik, 2008). The higher this number the more heat the material is able to conduct. In addition, copper can be used for the production of heat sinks because of its high thermal conductivity value of around 400 W/mK (Gallagher, et al., 1998). Its main disadvantage over aluminum is that it is three times heavier and more expensive.

Materials with high thermal conductivity (TC) and low coefficient of thermal expansion (CTE) are the choice for laptop computer heat sinks. A material suitable for this purpose must combine two basic properties: it must have a high thermal conductivity (TC) and a suitable coefficient of thermal expansion (CTE) (similar to semiconductors used in the manufacture of electronic circuits).

Carbon nanotubes (CNTs) with outstanding mechanical properties, very low thermal expansion (CTE≈0), and high thermal conductivity (Dai, 2002), are potential reinforcement materials for use in composites. According to theoretical predictions and experimental measurements, the thermal conductivity of CNTs reaches as high as 3000–6600 W/m/K (Kim, et al., 2001). Aluminum is one of the most important matrix materials for MMC.

So far, only a few studies have discussed the thermal conduction behavior of CNT/Al composites. Bakshi, et al., 2010 produced a composite of 10 wt.% CNTs/Al using plasma spraying and the thermal conductivity was only 25.4 W/m/K, much less than pure Al. Yamanaka et al. Yamanaka, et al., 2006, reported that the thermal conductivity of CNTs/Al composites decreased with increasing CNT content. The reported thermal conductivity for CNTs/Cu composites (Chu, et al., 2010 also showed a

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decreasing trend compared to pure Cu. The decrease in thermal conductivity was mainly related to the agglomeration of CNTs which resulted in the thermal resistance of the interface into the composite.

This research focuses on the thermal conduction behavior of aluminum matrix composites reinforced with multiwall nanotubes (MWNTs/Al composites) produced by the stir casting process. Stir casting processing is a very useful technique for bonding non-sinterable materials such as carbon nanotubes. In order to produce a homogeneous dispersion of carbon nanotubes in an aluminum matrix, the powdered carbon nanotubes were first modified before being incorporated into the aluminum melt.

2 MATERIALS AND EXPERIMENTALS PROCEDURES

2.1 Materials

Multiwalled carbon nanotubes supplied by Chengdu Organic Chemicals Co. Ltd., China (OD: 10 - 20 nm, length: 10 - 30 m and purity > 98%) were used in the present study. The colloidal palladium-tin activator made with a composition of 0.5 g palladium chloride (PdCl2), 50 ml 37% hydrochloride acid (HCl), 200 ml deionised water, 25 g stannous chloride. Cupric Sulphate Pentahydrate (98.5% Assay) and Sodium Carbonate Anhydrous (99.5% Assay) were supplied by Bofa Laboratotium. Sodium Hydroxide (99% Assay) was supplied by Bofa Laboratotium. Pottasium Sodium Tartrate Tetrahydrate or what is known as Rochelle salt (99% Assay) was supplied by Bofa Laboratotium. Cobalt (II) Chloride Hexahydrate (99% Assay) was supplied by Bofa Laboratotium. Formaldehyde 37% in aqueous solution was supplied by Bofa Laboratotium.

2.2 Experimental Procedures

The metallization of MWCNTs by copper was conducted in three steps. The process started by the activation of MWCNTs surface using colloidal Pd-Sn particles followed by the acceleration step to remove stannous hydroxide deposits on top of the activated surface. Finally, The electroless plating of Cu-Co on top of MWCNTs was performed as shown Figure. 1.

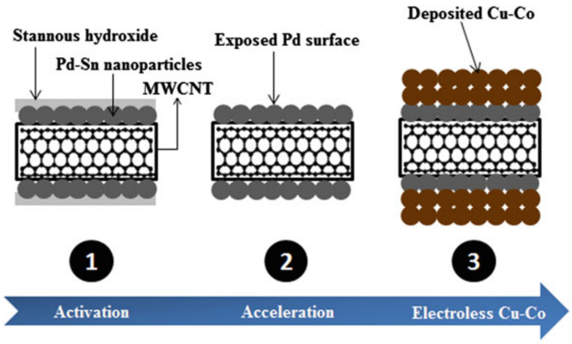

Figure 1: General scheme of Cu-Co electroless plating on MWCNTs.

In more detail, the procedure for coating WMCNTs with copper refers to the literature (Elsharkawi, 2018).

Figure 2 shows the color of the obtained copper coated MWCNTs powder.

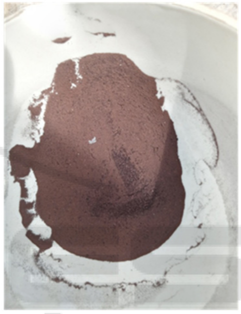

Figure 2: Filtered copper coated MWCNT's of a brown color.

The copper coated MWCNTs were characterized using scanning electron microscopy (SEM) analysis using (JEOL-JSM 6510 A) at the Materials Laboratory of Mechanical Engineering Udayana University.

2.2.1 Al-MWCNTs Composite Manufacturing Process

Materials for composites, aluminum and multiwall carbon nanotubes with varying compositions (0% ; 2%; 4%; 6%; 8% and 10% by weight MWCNTs) were included in the smelting kowi. Heated to a temperature of 700 oC with a time of 20 minutes and a stirrer speed of 200 rpm. The composite melt is poured into a cylindrical metal (steel) mold at room temperature.

2.2.2 Density of Al-Cu/MWCNTs Composites

The measurement of the density of the Al-Cu/MWCNTs composite material is a test object resulting from the stir casting process.

By knowing these quantities, the density of the Al-Cu/MWCNTs composite material can be determined using the equation (Birkeland, 1984),

$$\rho = \frac{m_s}{m_s - (m_g - m_k)} \; x \; \rho \; H_2O$$

with,

ρ = bulk density (gram/cm^3)
m_s = mass of the sample after drying in the oven (grams)
m_g = mass of sample suspended in water (grams)
m_k = mass of sample hanging wire (grams)
ρ H_2O = density of water = 1 gram/cm^3

2.2.3 Porosity of Al-Cu/MWCNTs Composites

By knowing these quantities, the porosity of the Al-Cu/MWCNTs composite material can be determined using the equation (Birkeland, 1984),

$$\rho = \frac{m_b - m_s}{m_b - (m_g - m_k)} \; x \; 100\%$$

with,

ρ = bulk density (gram/cm^3)
m_s = mass of the sample after drying in the oven (grams)
m_b = mass of the sample after soaking in water / saturated (grams)
m_g = mass of sample suspended in water (grams)
m_k = mass of sample hanging wire (grams)

2.2.4 Hardness of Al-Cu/MWCNTs Composite (Vickers Hardness Test)

The hardness of the Al-Cu/MWCNTs composite material was tested at the Metallurgical Laboratory of Mechanical Engineering, State University of Malang using a Microhardness Tester (ESEWAY, Model EW421AAT), and the test refers to the standard (Dowling, E.N., 1999); ASTM E 18 - 02.

Measure the diagonal length of each pressing result and the hardness value of the tested sample can be read directly on the microhardness tester monitor, perform at least 3 repetitions for each sample tested.

The hardness value of the Al-Cu/MWCNTs composite material can also be calculated using the following equation (Dowling, 1999).

$$VHN = \frac{2\,P}{d^2} \sin\frac{\alpha}{2} = 1{,}8564 \; \frac{P}{d^2}$$

with:

VHN = Vickers hardness value (kgf/mm^2)
P = pressing load (kgf)
d = average diagonal length (mm)
α = angle between diamond faces (136^o)

2.2.5 Thermal Conductivity

In this study, for the thermal conductivity test, Linear Heat Conduction Devices (TD1002a) were used.

Figure 3: Linear Heat Conduction Experiment (TD1002a).

The test object consists of Aluminum- MWCNTs composites with a thickness of 20 mm and a diameter of 30 mm

The energy that occurs in the heater is given by the equation:

$$W = V \times I$$

With ,

W = Electrical power (watts)
V = Voltage (volts)
I = Electric current (amperes)

Heat transfer that occurs:

$$\dot{q} = k.A.\frac{dT}{dx}$$

With ,

$\dot{q} = \frac{dQ}{dt}$= heat transfer rate (Watts)
K = conduction heat transfer coefficient (Watt/m.K)
A = Cross-sectional area of metal test object (m^2)
dT = temperature difference (K)
dx = distance between test points (T_1 and T_2)
for this case W = q

3 RESULTS AND DISCUSSION

3.1 Products of Al – MWCNTs Composites

The results of the Al – MWCNTs composite casting process with various compositions are shown in Figure 4.

Furthermore, the results of the Al - MWCNTs composite casting are formed (lathe process) into a diameter of 30 mm and a thickness of 20 mm, for the process of testing physical properties, morphological characterization and thermal conductivity, Figure 4.

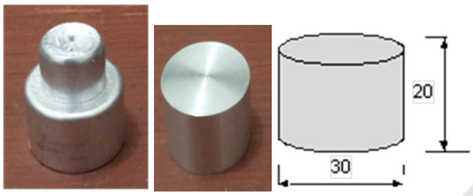

Figure 4: Al – MWCNTs composite casting product.

3.2 Physical Properties of Al –MWCNTs Composites

Based on the results of calculations using equation (1) for the density test, equation (2) for the porosity test, as well as testing the hardness properties of Aluminum – Multiwall Carbon Nanotube composites, which were carried out in the Lab. Metallurgy Mechanical Engineering, Udayana University, obtained data as shown in figure 5.

Figure 5: Physical Test Data for Al – MWCNTs Composites.

3.2.1 Effect of Reinforcing MWCNTs on Aluminum on Density

Based on Figure 6, it can be seen that with increasing MWCNTs content in Al – MWCNTs composites, the composite density tends to decrease. The lowest composite density of 2.535 g/cm^3 occurred when the MWCNTs content was 10% by weight. Meanwhile, the highest composite density of 2.754 gr/cm^3 occurred when the MWCNTs content was 2% by weight.

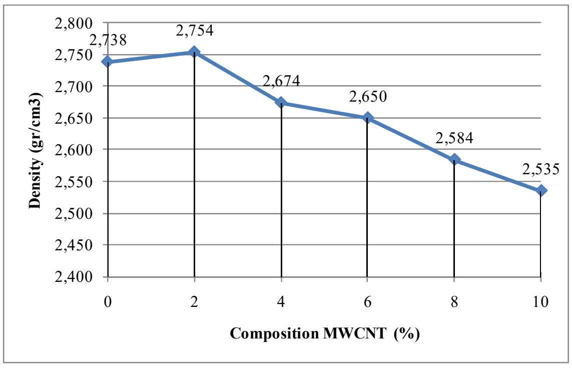

Figure 6: Graph of the relationship between density and composition.

3.2.2 Effect of Reinforcing MWCNTs on Aluminum on Porosity

Based on Figure 7, it can be seen that with increasing MWCNTs content in the Al – MWCNTs composite, the porosity of the composite tends to increase. The lowest composite porosity of 4.51% occurred when the MWCNTs content was 0% by weight. Meanwhile, the highest composite porosity of 9.96% occurred when the MWCNTs content was 6% by weight.

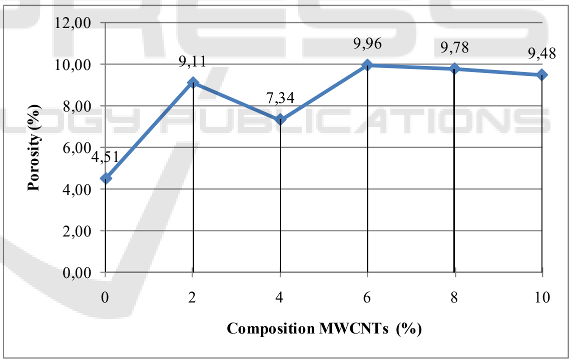

Figure 7: Graph of the relationship between porosity and composition.

3.3 Microstructure of Al-MWCNTs Composites

In the sample of the cast Al - Cu/MWCNTs composite, the Al grain morphology did not change much compared to Al fine, as shown in Fig. 3.4. This is because gravity casting is applied during the formation of the composite, which benefits the plasticizing of the powder to achieve full density. Grain boundaries are seen more clearly after repeated etching. In the pure Al samples, small grain growth

(Fig. 8a) and equiaxed grains were observed in all composites (Fig. 8b and 8c).

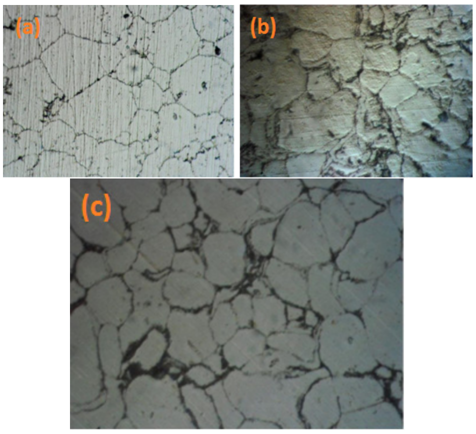

Figure 8: Microstructure image etched (a) pure Al, 1.0 wt.% (b) MWCNT/Al uncoated composite and (c) MWCNT/Al . Cu-coated composite.

In the composite sample, the MWCNTs were homogeneously dispersed at the grain boundaries and within the Al particles. By comparing the grain size of the composite samples (Fig. 3.5b and 3.5c) with the Al powder particles, it can be seen that the particle growth is very small. This is due to the embedding effect of MWCNTs which inhibits particle growth. It is important to note that the effect of Cu-coated MWCNTs increases more of the Al matrix binding interface. Therefore, small grain sizes were obtained in the Cu-coated MWCNTs/Al composites compared to the uncoated MWCNTs/Al composites, where little grain growth resulting in relatively larger grains was observed. Due to poor wettability in uncoated MWCNTs, the embedding of MWCNTs in Al particles was less during casting compared to Cu-coated MWCNTs/Al composites.

3.4 Effect of Reinforcing MWCNTs on Hardness

As previously mentioned, copper coated MWCNTs are added in varying percentages to the pure aluminum melt using casting techniques. The aim was to improve the wettability and dispersion between aluminum and MWCNTs which was reported to be very poor in a previous review (Agarwal, A., et al., 2011). The hardness of the sample was tested using a Vickers hardness tester. About 3 penetrations were carried out on a different area of each sample. The small variability of the Vickers hardness between the different indentations is indicative of the homogeneous distribution of Cu coated MWCNTs in the aluminum matrix. The results of the Vickers hardness number for various percentages of copper coated MWCNTs are presented in Table 1.

Table 1: Hardness test result data.

[redacted]

Based on Figure 9, it can be seen that with increasing MWCNTs content in Al – MWCNTs composites, the composite hardness tends to increase. The lowest composite hardness of 75 HV occurred when the MWCNTs content was 0% by weight. Meanwhile, the highest composite hardness of 91.3 HV occurred when the MWCNTs content was 10% by weight.

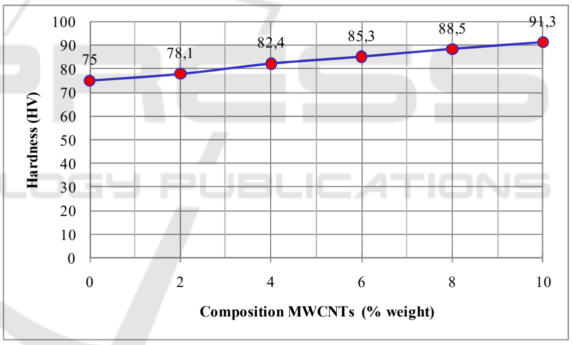

Figure 9: Graph of the relationship between hardness and composition.

It was found that adding 2, 4, 6, 8 and 10% copper-coated MWCNTs to pure aluminum resulted in an increase in Vickers hardness of 4.13 ; 9.87 ; 13.73 ; 18.00 and 21.73% are significant especially that, for example, 2% copper-clad MWCNTs have less than 0.1% of MWCNTs. This confirms the potential of the process used in producing good quality cast composites from Al-Cu/MWCNTs. Efforts are underway to optimize the casting process and to fully investigate the mechanical behavior of the composite.

3.5 Thermal Conductivity Test Results

For the thermal conductivity test, Linear Heat Conduction Devices (TD1002a) were used which was carried out in the Lab. The Basic Phenomenon of Mechanical Engineering, Udayana University. The size of the test object is 30 mm in diameter and 20 mm thick, with a power input of 50 Watt. Based on the test results and the calculation of the thermal conductivity of the Aluminum – Multiwall Carbon Nanotube composite, the data is obtained as shown in table 2.

Table 2: Data from the heat conductivity test.

[redacted]

Based on Figure 3.6, it can be seen that as the MWCNTs content increases in the Al-MWCNTs composite, the thermal conductivity of the Al-MWCNTs composite tends to increase. The lowest composite thermal conductivity of 252.42 W/m.K occurred when the MWCNTs content was 0% by weight. Meanwhile, the highest composite thermal conductivity of 442.32 W/m.K occurred when the MWCNTs content was 8% by weight.

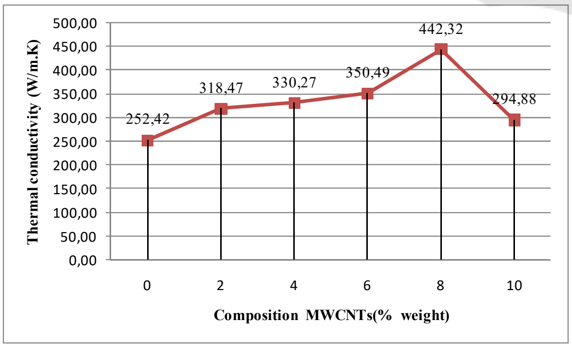

Figure 10: Graph of the relationship between thermal conductivity and composition.

4 CONCLUSION

In the process of making Al-MWCNTs composites with a stir casting process, it can be concluded that:

a. The higher the MWCNTs content, the density of the Al-MWCNTs composite decreased, while the porosity of the Al-MWCNTs composite increased.
b. The higher the MWCNTs content, the hardness and thermal conductivity of the Al-MWCNTs composite tend to increase.
c. The distribution of MWCNT in the aluminum matrix was uneven and agglomeration of MWCNT occurred at several locations.
d. Composites with the addition of <10 wt.% Cu/MWNTs have higher thermal conductivity than pure aluminum produced by the same liquid state processing.
e. The Cu/MWNTs/Al composites showed a maximum thermal conductivity of 442.32 W/m/K at 8 wt.% Cu/MWNTs. The increase in thermal conductivity is supported by the measured microhardness. The Cu/MWNTs/Al composites showed a maximum microhardness of 91.3 HV also at 10 wt.% Cu/MWNTs.
f. The results showed that the aluminum matrix composite reinforced with copper-coated multiwalled carbon nanotubes (Cu/MWNTs) is a potential material for high thermal conductivity applications.

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