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RESEARCH ARTICLE

The influence of layer thickness on the electrical property of metal-CNT (metal: Cu) composite

Salim Mustofa, Patricius Purwanto, Wisnu Ari Adi*

Center for Science and Technology of Advanced Materials (PSTBM) – BATAN, Kawasan Puspiptek Serpong, Tangerang Selatan 15314

* *Corresponding author: s_mustofa@batan.go.id*

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Abstract

The composite of Metal – MWCNT (Metal: Cu) were made by using solid-state reaction method for 1 hour at R.T. after mixing the Multiwalled - CNT (MWCNT) and Copper (Cu) powder with 3% weight of Cu. The result of electrical property measured using LCR meter indicated that the conductivities value of MWCNT/Cu was increased in proportion to the increase of layer thickness of composite and the increasing of frequency measurement. On the other hand, the capacitance value of the MWCNT/Cu composite sample was decreased by the increasing of frequency measurement. From the analysis of cole-cole plot, the MWCNT/Cu composite indicated the peak maximum at certainl frequency, which shows the possibility of achievable polarizability. We have measured the Raman spectra of MWCNT/Cu composites to evaluate the state of dispersion and the Cu-filler interactions reflected, by shifts changes of the peaks. All the Raman bands of the carbon nanotubes are seen at wave number around of 1326 cm^{-1} (D band), and a wave number around of 1617 cm^{-1} (second harmonic G band).

Keywords: MWCNT/Cu composites, layer thickness, conductivities, Raman spectroscopy, electrical property

INTRODUCTION

Carbon nanotubes (CNTs) have various types including Single-Walled Nanotubes (SWCNT), which are rolls of shorter graphite sheets that have a one-dimensional structure. Another form of CNT is Multiwalled Carbon Nanotube (MWCNT), which has a structure of more than one dimension and its size is short (V. Shanov *et al.*, 2006).

The solid materials have a conductivity property, whose value depends on frequency, geometric size, concentration and temperature, and additionally, at a certain temperature, a solid conductor material may be defective (K. J. Yulkifli *et al.*, 2009; T. Blanton *et al.*, 2011; P. R. Bandaru, 2007). From several papers, it was reported that CNTs are conductors that have better resistivity values than metals such as Cu at room temperature. The presence of defects or impurities on the CNT indicates the conductivity value of the CNT is lower than that of the defect-free CNT (Q. W. Li *et al.*, 2007). Carbon nanotube/Copper (CNT/Cu) thin film composites are fabricated by electrophoresis, electroplating methods and other techniques, which is used in interconnection applications (P. Liu *et al.*, 2008). One example of the application of nanocomposite materials of Cu/CNTs is for antimicrobial`s using (Sunil Kumar Singhal *et al.*, 2012).

In this study, an observation of MWCNT-Cu composite material fabricated by solid reaction or powder metallurgy was carried out. The characterization of the MWCNT-Cu composite was performed using Raman spectroscopy, and in addition to that, an electrical properties test was also conducted using the LCR meter. This study was done in order to determine the potential of MWCNT-Cu composite for materials with high thermal conductivity and other functional materials, as well as MWCNT-Al composite fabricated using a metallurgical method that showed excellent high thermal conductivity. Characterization by Raman Spectroscopy was used in this study because has a powerful technique that can be used to study the structure and electronic properties of carbon materials, including graphene/graphite and carbon nanotubes.

EXPERIMENTAL

The materials used in this research are Multi-Walled Carbon Nanotube (MWCNT) powder with purity > 95% and Copper powder (Cu) (Aldrich product), which has a 99.9% purity level. Cu and MWCNT powder are weighed with the following composition: MWCNT (97%) - Cu (3%). The total weight of powder mixture is 5 grams with thickness (1T) of Cu (3%) is around 1 mm (see figure 1). After pelletizing this powder mixture, then heated at a temperature of 900ºC for 1 hour. This kind of method is called as solid reaction or powder metallurgy process. The powder mixture which is the output of the solid reaction or powder metallurgy process is then characterized by Raman spectroscopy as well as electrical properties tested by LCR meter.

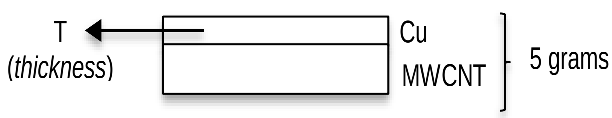

Fig. 1 The schematic illustration of the pellet sample of Cu/MWCNT composite fabricated by the solid reaction.

RESULTS AND DISCUSSION

Electrical ductivity

Electrical conductivity measurements were made using the HITESTER-3522-5 HIOKI LCR meter. Calculation of Cu-MWCNT electrical conductivity was carried out using the model equations performed by Padma Kumar et al, (P. Padma Kumar *et al.*, 2006) dan V.N. Bondarev et al (V.N. Bondarev *et al.*, 1999), that is :

$$\sigma(\omega) = \sigma_{dc} + A.\ \omega^{n} \quad (1)$$

This equation is then converted to the following logarithmic form:

$$Log\ \sigma(\omega) = log\ \sigma_{dc} + n.log\ A(\omega) \quad (2)$$

Where σ(ω) is AC conductivity (S/cm), σ_{dc} DC conductivity at frequency ω = 0 (S/cm), ω is frequency (Hz), n is the exponent fraction of Jonscher (0< n <1) and A is positive constant.

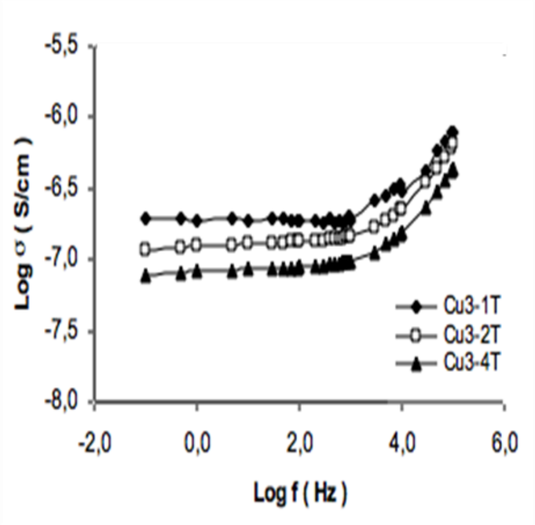

Fig. 2 The conductivity value of MWCNT/Cu composite with thickness variation (T = 1,0 mm).

Table 1 The conductivity of MWCNT/Cu.

Thickness (mm)	Conductivity σ_o (S/cm)	
	I	II
1,0	$1{,}92\times10^{-7}$	$1{,}91\times10^{-8}$
2,0	$1{,}23\times10^{-7}$	$1{,}03\times10^{-8}$
4,0	$1{,}23\times10^{-8}$	$6{,}96\times10^{-9}$

Table 2 The capacitance value of MWCNT/Cu.

Thickness (mm)	Capacitance value C_s (Farad)	
	I	II
1,0	$1{,}68\times 10^{-7}$	$1{,}30\times10^{-9}$
2,0	$5{,}82\times 10^{-7}$	$1{,}57\times10^{-9}$
4,0	$4{,}70\times 10^{-7}$	$1{,}04\times10^{-9}$

Fig. 2 shows that the conductivity value of MWCNT/Cu is increasing as the thickness concentration of Cu coating increases. In general, the conductivity of a material will increase when the material occurs a defect Frenkel or Shoctky (P. Padma Kumar *et al.*, 2006).

Calculation of conductivity value of MWCNT/Cu composite was done by using equation (2). The calculation results of the conductivity value are shown in Table 1, where it is seen that the conductivity value of the composite increases with the increase of Cu concentration into MWCNT. The rise in conductivity is due to the diffusion of Cu into MWCNT, which causes an increase in the number of charge carriers. The presence of Cu diffusion into MWCNT will decrease the activation energy required by the electrons to move from one lattice to the other, thus increasing the mobility of positive ions so that the conductivity will increase (V.N. Bondarev *et al.*, 1999). Other researchers have conducted research on the conductivity properties of frequency dependent, temperature, composition and phase transformation, where at a certain temperature, a material has a defect or phase transformation (Qingwen Li *et al.*, 2007; P. Liu *et al.*, 2008; Sunil Kumar Singhal *et al.*, 2012; V.N. Bondarev *et al.*, 1999), thereby giving the effect of increasing electrical properties such as conductance and others.

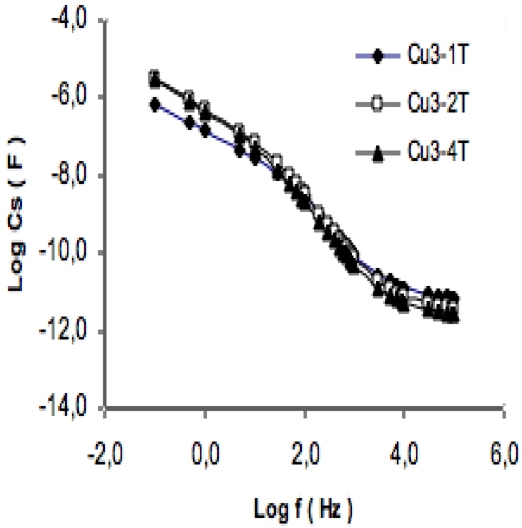

Fig. 3 The capacitance value of the MWCNT/Cu composite with thickness variation (T = 1,0 mm).

Fig. 3 shows that the capacitance curve is decreased as the frequency increases, and the capacitance value is increased as the Cu concentration increases. It is estimated that this increase in capacitance value is due to the increase of electron charge in Cu, while the decrease of capacitance value along with the increase of frequency is due to the charge saving power has reached saturation point. The calculation results of the capacitance value are shown in Table 2.

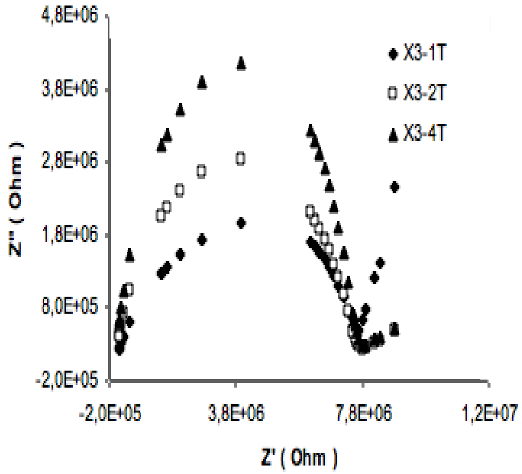

Fig. 4 The impedance value of the MWCNT/Cu composite with thickness variation (T = 1,0 mm).

The impedance curve showed in Fig. 4 is the maximum frequency for MWCNT/Cu composites with a frequency of f = 1 / (2π.R.C). From the analysis of the cole-cole plot of MWCNT/Cu, the peak point indicates the optimum point of a material working at a certain frequency.

Raman spectroscopy

The material was characterized by Raman spectroscopy using a 10 mV laser power and a wavelength of 768 nm.

Fig. 5a shows the Raman spectra of PVA and Fig. 5b is the Raman spectrum of the MWCNT/Cu3 composite material. The results of peak intensity analysis of D band and G band are shown in Table 3. An important characteristic of characterization using Raman spectroscopy to MWCNT is the derivation of the main spectrum of D band and G band (J.H. Lehman *et al.*, 2011).

Table 3 The conductivity of MWCNT/Cu.

Cu (%)	ID	IG	ID/IG
3	0,007857	0,006201	1,2670

Fig. 5 shows the D band's peak is at 1324.5 cm^{-1}, the second harmonic of G band`s peak is at 1604.5 cm^{-1} for the MWCNT/Cu composite.

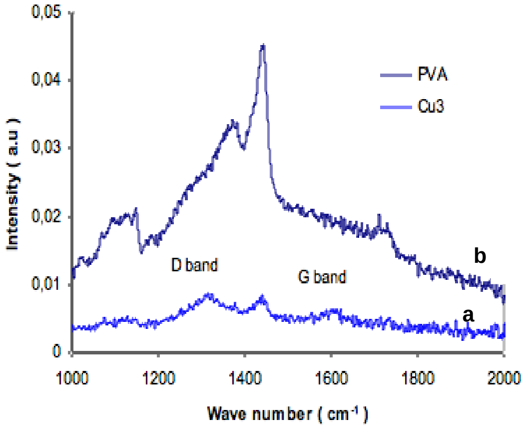

Fig. 5 Raman Spectroscopy of MWCNT-Cu. a). PVA b). MWCNT/Cu.

The peak of D band is enabled on the first sequence from the scattering process of carbon SP2 through its presence in vacancies, in the powder grain boundaries, or because of the presence of other defects, which lowers the symmetry of the lattice (E.F. Antunes *et al.*, 2007). The D band peaks are derived from disturbances inside the carbon Sp2 and can also arise due to lattice distortion on MWCNT. Therefore, the presence of D band peaks indicates a disruption in the MWCNT hexagonal framework (L. Bokobza *et al.*, 2012), which in this case is a disturbance of the mixing of Cu powder into MWCNT. The Raman intensity value decreases with increasing per cent weight of Cu in MWCNT, or in other words, the increasing impurity content that causing lattice distortion in the MWCNT hexagonal skeleton. The existence of the G band top itself corresponds to E2g mode (S. Costa *et al.*, 2008) of high-oriented graphite, and shows the presence of carbon crystals in the MWCNT sample. The Raman intensity value of the G band peak also decreases with the per cent decrease of Cu weight in MWCNT. The comparison of the peak intensity values of D band and G band (ID/IG) shown in Table 3 shows an increase along with the decrease of Cu per cent weight in MWCNT. The increasing of (ID/IG) indicates the increasing of crystallinity degree in MWCNT samples and indicates the decreasing of impurity levels in MWCNT. It means that the decreasing of ID/IG from the Raman spectra indicates the increase of capacitance, and in the other hand correspondent to the decreasing of the conductivity of the sample.

CONCLUSION

From the results of this study, it can be concluded that MWCNT/Cu composite with different Cu thickness indicates the conductivity of MWCNT/Cu composite is increased in proportion to the increase of Cu thickness, while the capacitance value is decreased. From the analysis of the cole-cole plot of the MWCNT/Cu composite plot, the peak point identifies the maximum point on a particular frequency. The Raman spectrum on MWCNT/Cu shows the intensity values of G and D bands decreased along with the increase in Cu. This result indicates the decrease of crystallinity degree of MWCNT/Cu and the increase of impurity level in MWCNT/Cu.

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