

## 다중벽 탄소나노튜브 표면개질 및 고분자 복합체 형태에 따른 열전도 특성 연구

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(2018년 2월 19일 접수, 2018년 4월 1일 수정, 2018년 4월 2일 채택)

## The Study of Multi-walled Carbon Nanotube Surface and Matrix Structure for Thermal Conductive Composite Material

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(Received February 19, 2018; Revised April 1, 2018; Accepted April 2, 2018)

**초록:** 고방열 복합소재를 위하여 고분자 경화 유무에 따른 두 가지 종류의 표면처리된 탄소나노튜브/알루미나/ETDS (epoxy-terminated dimethylsiloxane)복합체를 제작하였다. 탄소나노튜브는 산처리 후 실란커플링제 및 dodecylamine 을 이용하여 표면처리를 진행하였다. 실란커플링제와의 반응 메커니즘은 반응조건에 따라 조절되었다. 탄소나노튜브 표면처리에 따른 열전도도 특성 변화를 70 wt% 알루미나/ETDS 복합체를 기반으로 확인하였다. 탄소나노튜브 표면 처리의 효과는 복합소재의 종류에 따라 상이한 경향을 보이는데, 패드 형태의 복합소재에서는 아민기가 말단에 위치할 때 가장 우수한 열전도도 특성을 보이는 반면 그리스 형태의 복합소재에서는 긴 탄소사슬이 말단에 위치할 구조가 가장 높은 열전도 특성을 보였다. 또한, 실란올기가 말단에 위치할 때 탄소나노튜브의 복합화 가능한 함량이 가장 높아져, 표면처리 방법에 따른 탄소나노튜브의 최대 함량 및 열전도도 차이가 나타나는 것을 확인하였다.

**Abstract:** Two kinds of surface-modified MWCNT/Al<sub>2</sub>O<sub>3</sub>/ETDS composites, cured pad and uncured grease, were fabricated for high-thermal-conductivity materials application. MWCNTs were modified using aminopropyltriethoxysilane (APTES) and dodecylamine after acid treatment. The reaction mechanism of APTES and the surface structure of MWCNT was controlled using different reaction conditions. The effect of surface modification on the thermal conductivity was confirmed based on a 70 wt% Al<sub>2</sub>O<sub>3</sub>/ETDS composite. The thermal conductivity was found to be dependent on the composite type and surface structure of the modified MWCNTs; the amine-terminated MWCNTs showed the highest thermal conductivity among the pad type composites, while long carbon chain-terminated MWCNTs showed outstanding performance among the grease type composites. Moreover, the maximum processable MWCNT content was also influenced by the surface modification; structurally similar silanol groups affected the molecular mobility of the ETDS resin and they had the highest MWCNT content under the same process conditions.

**Keywords:** multi-walled carbon nanotube, surface modification, thermal conductivity, surface structure, matrix.

### Introduction

Multi-walled carbon nanotubes (MWCNTs) exhibit outstanding thermal and electrical properties, mechanical strength, and optical properties, and the development of mass production processes has led to an increase in the applications of MWCNTs in various fields not only academic but also indus-

trial, such as electronic devices, electrodes for energy storage devices, catalysts, and transparent devices.<sup>1-4</sup> Among them polymeric composite materials were considered one of the most promising application, consisting of small amounts of MWCNTs have been reported to exhibit enhanced thermal and mechanical properties.<sup>5</sup> To realize the true potential of MWCNTs, it is important to overcome their extreme hydrophobicity and high surface energy, as these factors lead to aggregation of MWCNTs in polar liquids and polymer matrices. Therefore, proper dispersion and good interfacial bonding between the CNTs and the polymer matrix are essential

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requirements for utilizing CNTs as effective reinforcements in polymer composites.<sup>6,7</sup> Many researchers have focused on surface modification of MWCNTs via functionalization or surface coating using various organic or inorganic materials to obtain highly dispersed MWCNT composites. These surface modification processes improve the intermolecular interactions, both chemically and physically, preventing aggregation of MWCNTs and ensuring uniform dispersion. However, an all-powerful surface modification method does not exist; the introduced surface structure and applied polymeric material hardly influence the properties of composite materials. Therefore, finding a suitable combination of surface modifications and matrices is challenging.<sup>8,9</sup> Generally, wet chemical oxidation using strong acid solutions is recognized as an efficient method for MWCNT purification, promoting dispersion and surface activation at the same time. The introduced functional groups enable further modification via chemical reactions at the MWCNT surface; it is the general starting point for proper surface modification with various matrices.<sup>10</sup>

Owing to miniaturization and multi-functionalization of electronic devices, MWCNTs have great application potential as high-thermal-conductivity materials. Especially, the development of light emitting diodes has accelerated research on high-thermal-conductivity materials to improve the lifespan and the reliability of the diodes. Most of the thermally conductive materials are composed of high-thermal-conductivity ceramic particles and polymeric materials.<sup>11,12</sup> Despite the ultra-high thermal conductivity of MWCNTs, their applications are very limited owing to their high electrical conductivity that causes leakage of electrical signals. However, thermally conductive materials containing small amounts of MWCNTs are promising because the nanotubes form heat conducting bridges between the other fillers. In this case, uniform dispersion of MWCNTs is very important as only a small amount of MWCNTs is used. The dispersed MWCNTs should not form aggregated bundles, instead they should connect with other particles of the matrix; therefore, appropriate surface modification is necessary.<sup>13,14</sup>

In this study, epoxy-terminated polydimethylsiloxane based thermally conductive composites were fabricated using  $\text{Al}_2\text{O}_3$  and MWCNTs. The MWCNTs were chemically modified using a strong acid solution to introduce oxygen functional groups, and the resulting MWCNTs were further modified using the silane coupling agent aminopropyltriethoxysilane (APTES) and dodecylamine. The APTES treatments were performed at two reaction conditions to control the surface struc-

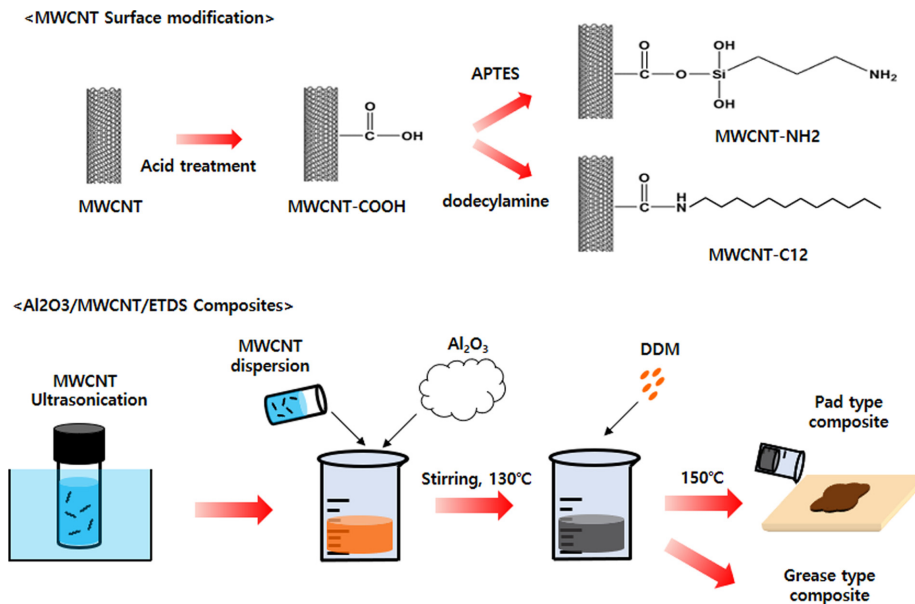
ture. The  $\text{Al}_2\text{O}_3$  concentration was fixed at 70 wt%, and the effect of surface modification of MWCNTs on the thermal conductivity was studied. Furthermore, two types of composite materials were fabricated: uncured grease type and thermally cured pad type with hardener. The thermal conductivity of the different composite materials was correlated with the surface structure of the MWCNTs and thermal curing.

## Experimental

**MWCNT Surface Modification.** The MWCNTs were purified and functionalized with carboxylic acid functional groups by heat treating the MWCNTs (4 g) in 400 mL of  $\text{H}_2\text{SO}_4$  and  $\text{HNO}_3$  (3:1 by volume) in an ultrasonic bath (Bransonic, MT-1510, 42 KHz) for 2 h at 25 °C to form a suspension. The suspension was then heated to 80 °C and stirred for 1 h. After the reaction, the acid solution was rinsed using deionized (DI) water and centrifugated. Finally, the carboxylic-acid-functionalized MWCNTs (MWCNT-COOH) were dried in a convection oven.

The prepared MWCNT-COOH was chemically modified using dodecylamine and APTES. The chemical reaction between MWCNT-COOH and APTES was carried out using two methods. In the first method, 1 g of MWCNT-COOH was dispersed in 400 mL of DI water using an ultrasonic bath. Next, 2 g of APTES was added, and the mixture was stirred at 80 °C for 6 h. The resulting particles (MWCNT-NH<sub>2</sub>) were rinsed with DI water, filtered three times, and then dried in a convection oven at 80 °C for 5 h to remove the solvent. In the second method, MWCNT-COOH was suspended in anhydrous dimethylformamide (DMF). Then, 2 g of APTES and 1 g of dicyclohexylcarbodiimide were added to the suspension and magnetically stirred at 80 °C for 6 h. The resulting particles (MWCNT-Si) were rinsed with DMF, filtered three times, and then dried in a convection oven at 100 °C for 5 h to remove the solvent. The reaction between MWCNT-COOH and dodecylamine (MWCNT-C12) was conducted via the same method used to prepare MWCNT-Si.

**Fabrication of  $\text{Al}_2\text{O}_3$ /MWCNT/ETDS Composites.** The uncured grease type and thermally cured pad type composite materials were prepared as follows. The grease specimen was fabricated using a simple mixture of filler and epoxy-terminated dimethylsiloxane (ETDS) resin. For this, the prepared surface-modified MWCNTs were initially suspended in ethyl alcohol (EtOH) under sonication for 3 h. Then, the prepared suspension was mixed with ETDS resin and  $\text{Al}_2\text{O}_3$  under mag-



**Scheme 1.** Fabrication process of Al<sub>2</sub>O<sub>3</sub>/MWCNT/ETDS composite.

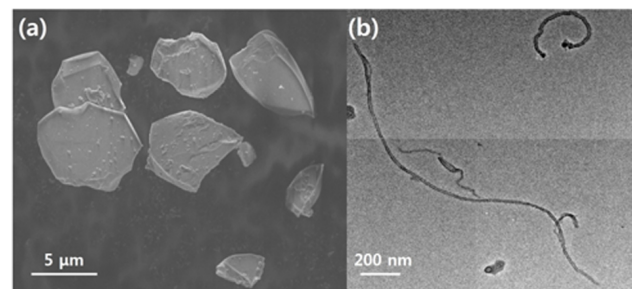
netic stirring at 130 °C to ensure homogeneous mixing and evaporation of the solvent. To fabricate the pad type composites, diaminodiphenylmethane (DDM) and ETDS resin were added in a 5:1 weight ratio and continuously mixed at 130 °C for 50 min. In this stage, the ETDS resin and the hardener reacted to form ETDS oligomers. Then, the mixture was poured onto a Teflon plate, and the remaining air bubbles were removed by degassing the mixture in a vacuum oven. Finally, the mixture film was cured at 150 °C for 3 h in a convection oven. The fabrication process of Al<sub>2</sub>O<sub>3</sub>/MWCNT/ETDS composite was shown in scheme 1.

**Characterization.** The fabricated surface-modified MWCNTs were characterized by X-ray photoelectron spectroscopy (XPS, Thermo U.K. K-Alpha) using an Al K $\alpha$  X-ray source (1486.6 eV) and a hemispherical analyzer. During curve fitting, the Gaussian peak widths were constant in each spectrum. Thermogravimetric analyses (TGA; TGA-2050, TA Instruments) of the samples were carried out to examine the thermal degradation process. Four-milligram samples were heated to 800 °C at a heating rate of 10 °C min<sup>-1</sup> under nitrogen and air atmospheres. Field emission scanning electron microscopy (FE-SEM, Sigma, Carl Zeiss) and high-resolution transmission electron microscopy (HR-TEM, JEM-3010) were used to examine the morphology of the fabricated MWCNTs and the composites. The thermal transport performance of the cured composites was characterized by laser flash analysis (LFA, Netzsch Instruments Co., Nanoflash LFA447) and differential

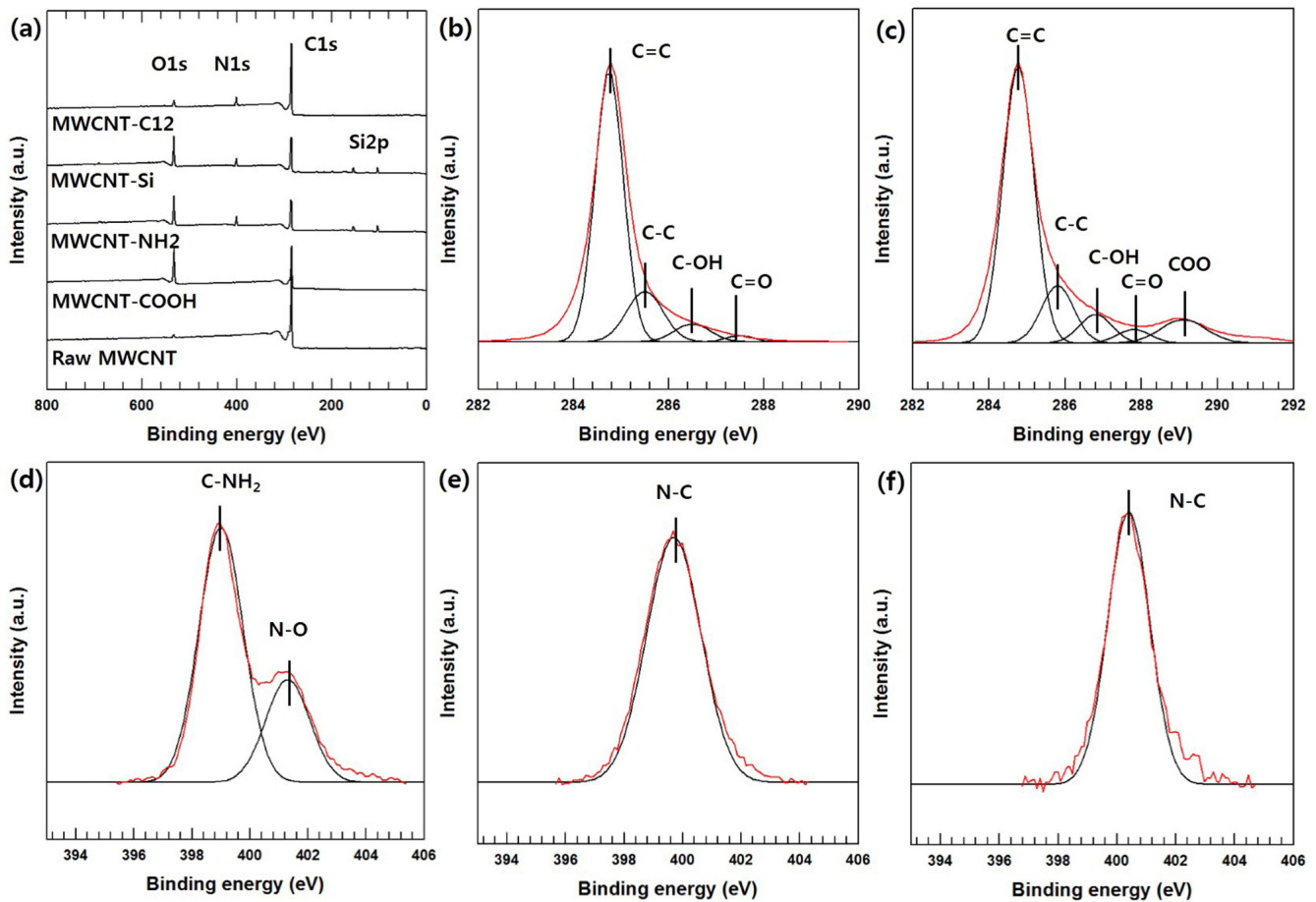
scanning calorimetry (DSC, Perkin-Elmer Inc., DSC-7) at 25 °C. The transferred signal initiates a thermal equilibration process in the composite specimen, which is recorded using a difference detector at the rear surface and then used to evaluate the thermal diffusivity. The bulk densities  $\rho_{\text{comp}}$  (g cm<sup>-3</sup>) of the specimens were measured using the Archimedes water displacement method. The thermal conductivity ( $k$ ) was calculated by multiplying the thermal diffusivity, density, and specific heat capacity of the composite. The thermal conductivity of the uncured composites was measured by laser flash analysis (LFA, Netzsch Instruments Co., Nanoflash LFA457) using platinum holder for liquid specimen.

## Results and Discussion

The morphologies of used thermal conductive fillers were observed via FE-SEM and HR-TEM which are shown in Fig-



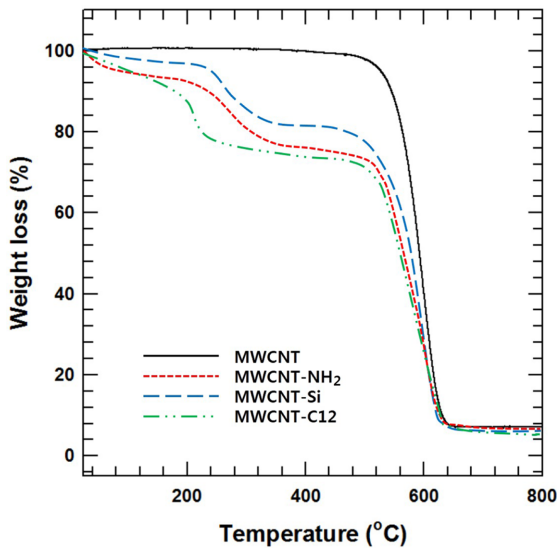
**Figure 1.** Morphology of used (a) Al<sub>2</sub>O<sub>3</sub>; (b) MWCNT.



**Figure 2.** XPS analysis of MWCNT and surface modified MWCNTs. (a) wide scan spectra; (b)-(c) C1s spectra of raw MWCNT and MWCNT-COOH; (d)-(f) N1s spectra of MWCNT-NH<sub>2</sub>, MWCNT-Si and MWCNT-C12.

ure 1. In this study, Al<sub>2</sub>O<sub>3</sub> was used main filler and small amount of MWCNTs were added. The mean diameter of Al<sub>2</sub>O<sub>3</sub> 5 microns. And, the length of the MWCNTs is approximately few microns, and their diameter is 20 nm. The surface modification of MWCNTs via acid treatment and further chemical modification was confirmed via XPS analysis. Figure 2(a) shows the wide scan spectra of pristine MWCNTs and the surface-modified MWCNTs. It is known that MWCNTs are composed of mostly carbon atoms and some oxygen atoms; a high-intensity peak corresponding to carbon (~285 eV) and a low-intensity peak corresponding to oxygen (~530 eV) are observed in the XPS spectra. The intensity of the oxygen peak increases considerably after acid treatment, indicating the introduction of significant amounts of oxygen functional groups. Furthermore, the additional low-intensity peak observed at approximately 100 and 400 eV in the spectra of chemical modified MWCNTs is attributed to APTES and

dodecylamine containing silicon and nitrogen atoms. To further confirm the chemical reactions, the C1s and the N1s spectra of the MWCNTs were deconvoluted, and their results are shown in Figures 2(b)-(f). As shown in the C1s spectra, the pristine MWCNTs are composed of C-C and C-O bonds, which correspond to hydroxyl and ketone groups, respectively. After the acid treatment, the number of oxygen functional groups increases, and carboxyl groups are formed because of further oxidation of existing oxygen functional groups. The difference between MWCNT-NH<sub>2</sub> with MWCNT-Si and MWCNT-C1<sub>2</sub> is clearly apparent in the N1s spectra; MWCNT-NH<sub>2</sub> shows the presence of amine groups while MWCNT-Si and MWCNT-C1<sub>2</sub> shows the presence of amide groups. Similar results were obtained for MWCNT-C1<sub>2</sub>, because MWCNT-C1<sub>2</sub> and MWCNT-Si are formed from similar reaction mechanisms. These results indicate that the reaction site can be controlled by the reaction conditions. In addition, the

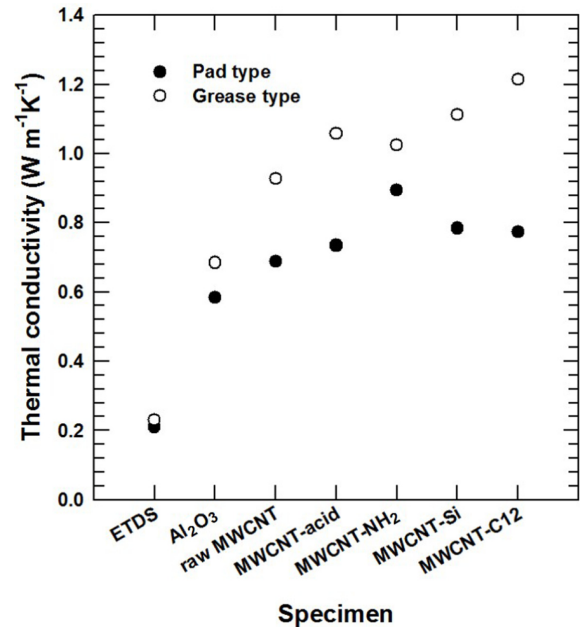


**Figure 3.** TGA analysis for MWCNT and surface modified MWCNTs.

XPS spectrum of MWCNT-Si and MWCNT-C12 shows a single amide peak, which confirms the chemical reaction between MWCNTs and APTES, dodecylamine without the formation of unreacted impurities.<sup>15,16</sup>

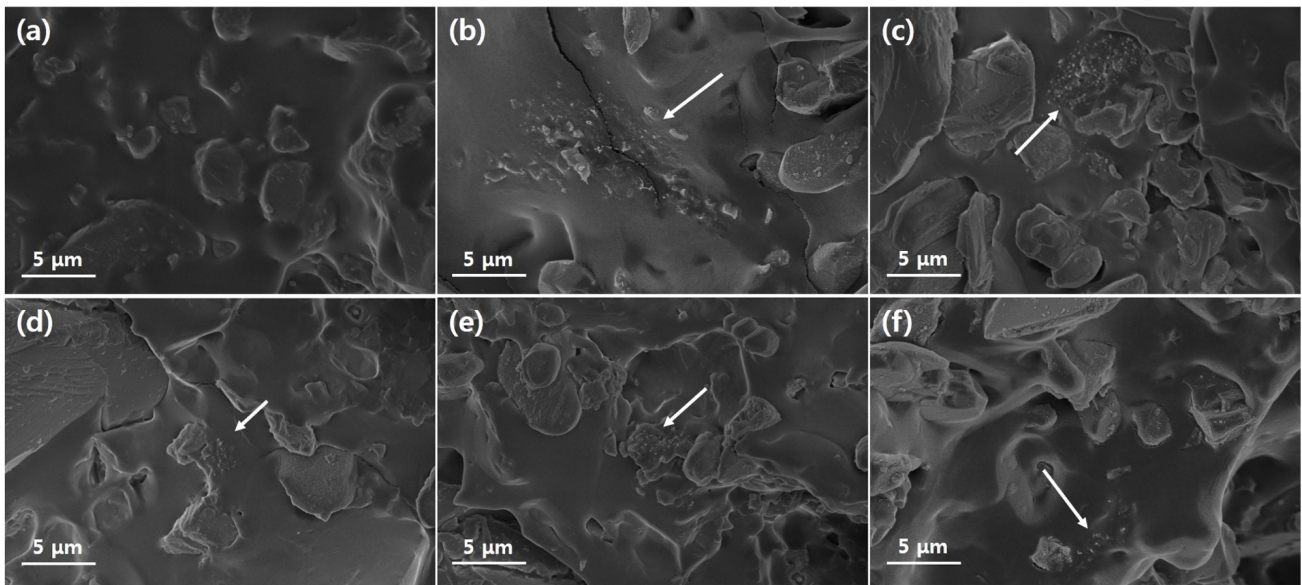
The quantitative analysis of the surface-modified MWCNTs was performed using TGA, and the results are shown in Figure 3. In air atmosphere, most of the MWCNTs were burnt at approximately 550 °C, and the weight losses of surface-modified MWCNTs occurred at approximately 250 °C, which indicate thermal degradation of APTES and dodecyl amine. The weight ratios of the three types of surface-modified MWCNTs are similar, approximately 23, 19, and 24 wt%, owing to the similar molecular weights of the introduced materials.

The thermal conductivity of the Al<sub>2</sub>O<sub>3</sub>/MWCNTs/ETDS composite was determined as a function of the surface modification process (Figure 4). Each composite type consisted of a fixed weight fraction of total filler contents, and the MWCNT content was fixed at 70 and 0.3 wt%. In both the cases, the addition of MWCNTs significantly enhanced the thermal conductivity. These results are in accordance with previously reported results, confirming that the ultra-high thermal conductivity MWCNTs can form thermally conductive bridges between the Al<sub>2</sub>O<sub>3</sub>-Al<sub>2</sub>O<sub>3</sub> interfaces. Furthermore, the surface-modified MWCNTs exhibited a higher enhancement ratio because the introduced functional groups or molecules enhance the dispersibility of the MWCNTs in the composite. Interestingly, all the grease type specimens exhibited higher thermal conductivity than the pad type specimens, owing to shrinkage during



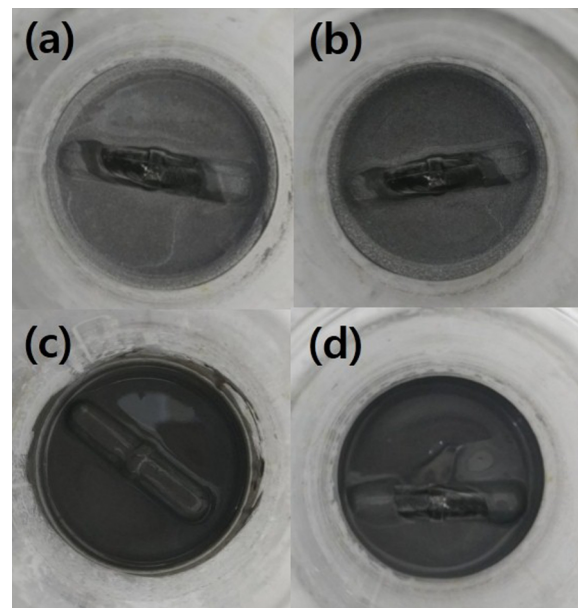
**Figure 4.** Thermal conductivity of composites with two types of composites.

thermal curing. In general, thermoset resins exhibit a shrinkage of approximately 5 vol% during curing, which results in air voids between the filler-matrix interfaces.<sup>17</sup> The air voids are not only thermally insulating, but also interrupt the heat flow pathway. The thermal conductivity was found to depend on the composite type and the surface modification process; MWCNT-NH<sub>2</sub> exhibited outstanding performance among the cured composites, while MWCNT-C12 exhibited the highest thermal conductivity among the uncured grease type composites. These behaviors are attributed to two reasons: structural differences between the polymeric matrices and reactions during thermal curing. The interfacial interaction is significantly influenced by the molecular structure of both the materials. In particular, the interfacial affinity is influenced by polar and unipolar factors. The polar factors include dispersion force, polarity, hydrogen bonding, induction, and acid-base components, and the nonpolar factors include the structural similarity effect and molecular chain entanglement.<sup>18,19</sup> The structure of the uncured ETDS resin is almost linear, with a dimethylsiloxane backbone and terminal epoxide groups. Therefore, it exhibits an almost nonpolar behavior, and it can be easily entangled with long carbon chains such as dodecylamine at the surface of MWCNTs, resulting in MWCNT-C12 has outstanding thermal conductivity among the grease type composites. The structure of MWCNT-Si is similar with that



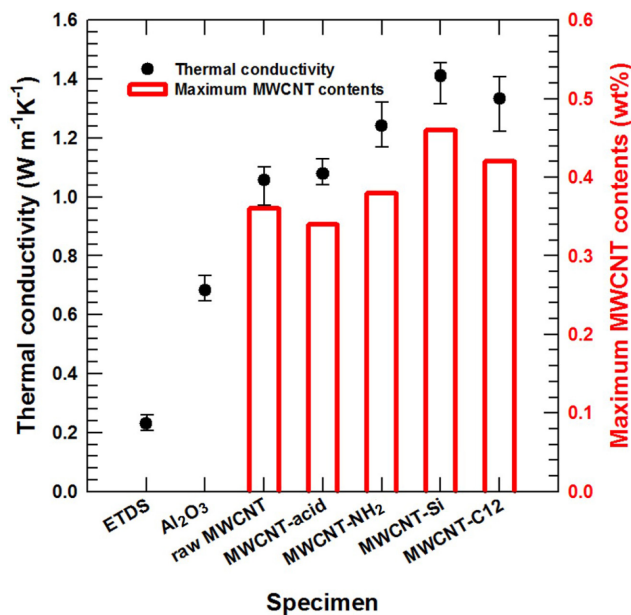
**Figure 5.** Cross-sectional SEM images of pad type composites. (a)  $\text{Al}_2\text{O}_3$  composite; (b) raw MWCNT composite; (c) MWCNT-COOH composite; (d) MWCNT- $\text{NH}_2$  composite; (e) MWCNT-Si composite; (f) MWCNT-C12 composite (the arrow mark denotes the aggregated MWCNTs).

of un-cured ETDS resin at the surface; therefore, its composite exhibits a higher thermal conductivity than the other MWCNT composites. On the other hand, the pad type composite after curing of the ETDS resin exhibits a different behavior; the amine group of MWCNT- $\text{NH}_2$  can undergo chemical reactions with the epoxide group of the ETDS resin, resulting in chemical linkages between the MWCNTs and the polymeric matrix, thus exhibiting high thermal conductivity. Further evidence for the enhancement in the thermal conductivity was obtained via cross-sectional FE-SEM studies of surface-modified MWCNTs. As shown in Figure 5(a), large-sized aggregated MWCNT bundles are observed in the pristine MWCNT composite. However, the size of the aggregated MWCNT bundles is significantly decreased after acid treatment. This indicates that a simple solution treatment and the resultant functional group introduction also influence the dispersibility of the MWCNTs. These results correlate well with the thermal conductivity behavior. However, dispersed MWCNTs or MWCNT bundles are not observed on the chemically modified MWCNT composites. Owing to the extremely small size of MWCNTs and the enhanced interfacial affinity with the polymeric matrix, the matrix was almost fully covered with the MWCNTs and the extruded filler was not observed. These results also demonstrate the enhancement in the thermal conductivity. Unfortunately, the dispersion state of the MWCNTs in grease type composite materials could not be observed by



**Figure 6.** MWCNT dispersion states on  $\text{Al}_2\text{O}_3/\text{ETDS}$  mixture: (a) raw MWCNT; (b) MWCNT- $\text{NH}_2$ ; (c) MWCNT-Si; (d) MWCNT-C12.

FE-SEM, as liquids cannot be observed using electron microscopy. Therefore, to compare the MWCNT dispersion state in ETDS resin, the optical images of the  $\text{Al}_2\text{O}_3/\text{MWCNTs}/\text{ETDS}$  mixture were recorded, which are shown in Figure 6. In the case of pristine MWCNTs, the mixture has a bright grey color (Figure 6(a)).



**Figure 7.** Maximum MWCNT contents and their thermal conductivity.

However, the surface-modified MWCNT composites are darker in color; especially, the MWCNT-Si composite is almost black in color. The bright color is attributed to the presence of white Al<sub>2</sub>O<sub>3</sub> particles in the inhomogeneously dispersed MWCNT composite, as aggregated bundles cannot absorb the whole visible radiation. On the other hand, the surface-modified and well-dispersed MWCNTs effectively interrupt light scattering, and hence, the mixture was darker or black in color. The dispersion states correspond well with the thermal conductivity behavior of the grease type specimens.

In the grease type specimens, increasing amounts of MWCNTs were added at fixed Al<sub>2</sub>O<sub>3</sub> contents, until the mixture attained the maximum processable MWCNT content under the same conditions. The maximum processable MWCNT contents and their thermal conductivity are shown in Figure 7. It is evident that the maximum processable filler contents depend on the surface structure of MWCNTs. The processability of the fluidic mixture was strongly influenced by the viscosity of the mixture, and the viscosity is closely related with the surface area and the surface affinity.<sup>20,21</sup> The surface areas of the surface-modified MWCNTs are assumed to be almost same because the thin coating layer has a negligible influence on the surface area. However, the surface interaction between the MWCNTs and ETDS resin has a significant influence on the viscosity. It is well known that an increase in the filler content increases the viscosity of the mixture, because

some of the resin covering the filler loses its chain mobility. Especially, strong chemical interactions, such as strong reactive functional groups, attract the surrounding resin and increase the viscosity, restricting the maximum permissible MWCNT contents. However, the long carbon chain of MWCNT-C12 could only entangled resin, its composite has less influenced on polar factor. On the other hands, the surface structure of MWCTN-Si is very similar to that of the ETDS resin; any attractive force for restriction of chain did not conducted during mixing process, resulting in MWCNT-Si composite showing the highest processable MWCNT content. The maximum filler contents strongly affect the thermal conductivity; higher MWCNT contents increase the thermal conductivity under the same compositing process. The MWCNT-Si composite exhibited the highest thermal conductivity among the composites based on same Al<sub>2</sub>O<sub>3</sub> contents.

## Conclusions

In this study, surface modifications of MWCNTs and their composite materials were carried out to develop high-thermal-conductivity materials. The MWCNTs were functionalized using a H<sub>2</sub>SO<sub>4</sub>/HNO<sub>3</sub> solution to introduce oxygen functional groups. After the functionalization, further surface modification was performed using APTES and dodecylamine. The APTES treatment was conducted using two kinds of solvents, EtOH and anhydrous DMF with DDM catalysis. APTES was hydrolyzed using EtOH, and the resulting hydroxyl groups reacted with MWCNTs. However, the amine groups of APTES reacted with MWCNTs in the case of anhydrous DMF. The surface modification of the MWCNTs using various reactants and reaction conditions was confirmed via XPS analysis. To verify the effect of MWCNT surface modification, composite materials containing 70 wt% Al<sub>2</sub>O<sub>3</sub> based on ETDS matrix were fabricated, and their thermal conductivities were studied. Two types of composites were prepared: thermally cured pad type with additional hardener and uncured grease type. Among the pad type composites, MWCNT-NH<sub>2</sub> showed the highest thermal conductivity as the amine group could react with the epoxide group of ETDS resin. Among the grease type composites, MWCNT-C12 showed the highest thermal conductivity as the long carbon chain could be easily entangled with the linear structured ETDS resin. Furthermore, higher amounts of MWCNT were processable in MWCNT-Si composites compared to the other MWCNTs under the same experimental conditions; this also resulted in a higher thermal

conductivity. Thus, the MWCNT surface was modified appropriately according to the matrix to achieve a high thermal conductivity.

**Acknowledgments:** This Research was supported by the Chung-Ang University Research Grants in 2017 and was also supported by the Human Resources Development (No.20184 030202070) of the Korea Institute of Energy Technology Evaluation and Planning (KETEP) grant funded by the Korea government Ministry of Trade, Industry and Energy.

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