Mechanical, electrical, piezoelectric and electro-active behavior of aligned multi-walled carbon nanotube/cellulose composites

Sungryul Yun, Jaehwan Kim *
Center for EAPap Actuator, Dept. of Mechanical Engineering, Inha University, 253 Yonghyun-Dong, Nam-Ku, Incheon 402-751, South Korea

ARTICLE INFO
Article history:
Received 12 March 2010
Accepted 26 September 2010
Available online 1 October 2010

ABSTRACT
Multi-walled carbon nanotubes (MWCNTs) were covalently grafted to cellulose to make an MWCNT/cellulose (M/C) composite. Aligned M/C composite was obtained by mechanical stretching process. The stretching effect was demonstrated by observing morphology as well as measuring mechanical, electrical and piezoelectric properties of the M/C composite. The influence of aligned MWCNTs on the actuator performance of the M/C composite was evaluated in terms of bending displacement and resonance frequency depending on the stretching ratio and environmental humidity level. The aligned MWCNTs contributed to remarkably enhancing the mechanical and piezoelectric properties, but also improving actuator performance of the M/C composite.

1. Introduction
Cellulose is one of the most naturally abundant biopolymers. Due to its biocompatible and biodegradable characteristics, application fields of cellulose have been enlarged from textile to immobilization of proteins, antibodies and pharmaceuticals [1]. Since electro-mechanical coupling effect in wood was discovered in early of 1960s [2], it has been known that piezoelectric response in wood is strongly associated with shear piezoelectricity of oriented cellulose crystallites [3]. However, despite these early studies, the potential of cellulose as a smart material that can be used for sensors and actuators has not been fully explored. Recently, cellulose paper has been re-discovered as a smart material that can be utilized for sensors and actuators [4]. This smart material is termed as electro-active paper (EAPap) [5]. EAPap has many advantages in terms of lightweight, dryness, low cost, biodegradability, large deformation and low power consumption. However, EAPap needs to overcome its drawbacks of low output force and low actuating frequency band.

Carbon nanotube (CNT) has unique electrical, mechanical, and thermal properties [6]. CNT has been utilized to improve mechanical property of polymers [7–9]. By introducing small amount of CNTs to polymers using mechanical blending or various chemical grafting methods, the CNT/polymer composites improved their elastic modulus and mechanical strength [10–13]. Since many researchers gave much effort to manipulate electrical properties of polymers by introducing CNTs in polymer matrix, the CNT has been recognized as a fascinating material to convert from dielectric polymer to conductor [14,15], to improve electrochemical response for polymer actuator [10,16,17] and to enhance dipolar alignment in piezoelectric polymer [18]. To realize reproducible high-performance and multi-functionality of CNT/polymer composites for practical applications, alignment of CNTs in or on polymer matrix has been proposed by stretching [19], spin coating [20], electric field [21] and magnetic field [22]. The aligned CNTs remarkably contributed to utilizing CNT/polymer composites to practical sensor applications. However, most of researches for the aligned CNTs have been

* Corresponding author: Fax: +82 32 8327325.
E-mail address: jaehwan@inha.ac.kr (J. Kim).
0008-6223/$ - see front matter © 2010 Elsevier Ltd. All rights reserved.
doi:10.1016/j.carbon.2010.09.051
focused on sensor applications. Only a few groups gave their attention to discover unveiled potential functionalities of the aligned CNTs by studying infrared actuation [19,23].

In our previous research, multi-walled carbon nanotubes (MWCNTs) have been introduced to cellulose by mechanical blending as well as covalent grafting to overcome drawbacks of cellulose EAPap [24–26]. The implementation of homogeneous distribution of MWCNTs in cellulose matrix with the covalent grafting allows to construct stable electron pathways for paper transistor [27], and to reinforce the mechanical property for EAPap actuator as compared to mechanical blending method [24,25]. The reinforced mechanical property increased its resonance frequencies of cellulose EAPap to higher band. However, except for the roles of CNTs in reinforcing the mechanical and electrical property of cellulose, we have been given less attention to other functionality of the CNTs that can be realized by not only homogeneous distribution of CNTs, but also alignment of the CNTs covalently grafted to cellulose, which can be a piezoelectric polymer. In-depth study for partially unveiled potential of CNTs to enhance dipolar alignment as filler in piezoelectric polymer matrix may contribute to opening routes to develop advanced piezoelectric polymers with high-piezoelectric response.

Thus, in this study, we investigated not only the presence of covalent grafting of MWCNTs on the cellulose, but also influence of aligned MWCNTs in MWCNT/cellulose (M/C) composite in terms of mechanical, electrical, morphological and piezoelectric properties of the aligned M/C composite. The actuator performance of the aligned M/C composite is evaluated by measuring its bending displacement and resonance frequency depending on the stretching ratio and the humidity level.

2. Experimental

2.1. Fabrication process

The cotton pulp (Buckeye) with the degree of polymerization (DP), 4500 and lithium chloride (LiCl) (Junsei Chemical) was dried in an oven at 100 °C to evaporate absorbed water molecules. The cotton pulp was mixed with LiCl/anhydrous N,N-dimethylacetamide (DMAc) (Sigma–Aldrich) in proportion to cotton cellulose pulp/LiCl/DMAc (2/8/90). The cellulose was dissolved in the solvent system by heating at 155 °C with mechanical stirring followed by the solvent exchange technique [28].

MWCNTs (Aldrich) with a nominal outside diameter of 10–30 nm, average length of about 20 μm and purity more than 95% were functionalized with carboxyl and hydroxyl groups by nitric acid treatment [29], filtered, and washed with deionized (DI) water until the pH reached to 7. The functionalized MWCNTs (6 mg) were dispersed in DMAc (10 ml) using a sonicator operating with a power of 40 W at 40 kHz (Hwashin Tech., Powersonic 603) for 10 min. N,N-Carbonyldiimidazole (CDI) (120 mg) was dissolved in DMAc (10 g). The solutions were mixed and sonicated in the ultrasonic bath at 60 °C for 12 h. The process contributed to convert carboxyl groups on MWCNTs to imidazolide. By reaction of imidazolide–MWCNTs (1.5 wt%) dispersed solution with cellulose solution at 60 °C for 18 h, MWCNTs were covalently grafted to cellulose chains. The weight percent of MWCNTs was defined by calculating weight percent of MWCNT–imidazolide (6 mg) with respect to that of cellulose (400 mg) included in cellulose solution (20 g). Fig. 1 represents the scheme of the reaction process between MWCNTs and cellulose. The MWCNTs covalently grafted cellulose (M/C) solution was spin-coated on a silicon wafer, and cured in three steps; isopropyl alcohol (IPA), deionized (DI) water/IPA mixture (the ratio of 4:6) and DI water in sequence. This slow curing process was performed to prevent instant aggregation of MWCNTs and to eliminate remnant Li+(DMAc)x macrocations as well as solvents [30].

To align MWCNTs in the M/C composites, both end of the wet M/C composites with the standardized size of 10 cm (length) × 10 cm (width) were fixed on a stretching machine and mechanically stretched with 10% strain/min. During the stretching process, the samples were dried for 2 h by exposing to a near infra-red ray heater of 80 °C. The ratio of width shrinkage was increased 0.1–0.55 as stretching ratio increased from 1.0 to 1.8. After the process, morphology of the sample changed from squared shape to dogboned one.

2.2. Chemical and morphological characterization

Element analysis was performed by element analyzer (Thermo Scientific, EA1112) to quantitatively determine chemical composition of MWCNTs functionalized with carboxyl group and imidazolide, respectively. Infra-red (IR) absorption and
Raman spectra of M/C composites were taken by Fourier transform infra-red (FT-IR) vacuum spectrometer (VERTEX, 80 V) and Raman spectrometer (Bruker, RFS 100/S) with YAG laser (wavelength: 1064 nm) to qualitatively demonstrate the covalent grafting of MWCNTs to cellulose chains. Thermo-gravimetric analyzer (TGA) (NETZSCH, STA 409 PC) was utilized to investigate the thermal weight loss of samples. The samples were analyzed by burning 10–15 mg in air environment with heating up to 700 °C and heating rate (20 °C/min). Transmission electron microscopy (TEM) observation was performed by TEM (Philips, CM200) to investigate the dimensional change that can be occurred by reaction to produce functionalized MWCNTs. The samples was prepared by casting samples dispersed in acetone on a carbon coated TEM grid (IGC 200, PELCO®), and dried in a vacuum oven at 40 °C for 12 h. The morphological change induced by alignment of MWCNTs in the M/C composites was investigated using a scanning electron microscope (SEM) (Hitachi, S-4000) at an acceleration voltage of 15 kV. The samples were prepared by coating platinum layer using ion sputter (EMITECH, K575X).

2.3. Mechanical and electrical characterization

The effect of aligned MWCNTs on the mechanical and electrical properties of the composites was evaluated by measuring Young’s modulus and electrical resistance, respectively. Young’s modulus was obtained in an ambient condition (20 %RH) according to the ASTM D-882-97 as a standard test method for tensile elastic properties of thin plastic sheeting. Samples for the tensile test were prepared with the size of 7 cm (length) × 1 cm (width). Electrical resistance was measured as a function of frequency ranging from 20 Hz to 10 kHz using LCR meter (HP, 4283A) integrated with probe station. To measure the property in aligned direction of the M/C composite, two rectangular electrodes (Au) with a constant distance (50 μm) were patterned on the composites by lift-off process.

2.4. Piezoelectric characterization

Dipolar polarization behavior of aligned M/C composites was studied by a thermally stimulated current (TSC) measurement, which analyzes depolared current profile with each function of temperature and electrical poling field. Based on the polarization behavior, piezoelectric response of the aligned M/C composite was investigated by measuring piezoelectric charge constant [d31]. Before performing the TSC measurement, a glass transition temperature (Tg) that normally used as a temperature for dipolar polarization of piezoelectric polymer was obtained by a differential scanning calorimetry (DSC) (NETZSCH, DSC 200F3). The samples for DSC analysis was prepared by cutting M/C composites into pieces, and their heat flows were analyzed by heating 3 mg of the pieces from room temperature to 350 °C in air with heating rate of 30 °C/min.

For TSC measurement, a classical procedure which is composed of four steps was performed consecutively. The procedure includes (1) heating the sample to Tg (165 °C), (2) applying the electric field at Tg for 5 min, (3) cooling the sample down to 25 °C with the field onto freeze-in dipolar alignment, (4) reheating the sample at a slow rate using furnace while monitoring the depolared current to quantify the dipolar alignment that took place in step 2. Samples for the TSC measurement were prepared by coating gold electrode on both sides of the M/C composites with the size of 1 cm (length) × 1 cm (width). A function generator (33220A, Agilent) with high voltage power amplifier (PZD 350 M/S, TREK) was used to apply the electric field ranging from 500 kV/m to 1.5 MV/m. A Picoammeter (6485, Keithley) collected the depolared current of the sample depending on the temperature that is modulated by the furnace with the constant heating rate of 0.1 Ks⁻¹. Overall sensitivity of the measurement system is about 2.0 pA and 10 mK in the current and the temperature measurement, respectively.

To quantify the piezoelectricity, induced charges of the M/C composites were measured by quasi-static method [31,32]. The experimental setup for induced charge measurement consists of a pull test machine, an environmental chamber and picoammeter (Keithley, 6485). In the pull test machine, load cell (Daecell Korea, UU-K010) and linear scaler (Sony Japan, GB-BA/SR128-015) were used to measure the applied load and displacement. M/C composites on which gold electrodes were coated on both sides were installed in the pull test machine by gripping the samples with the ASTM standard grip for polymer film tests. The electrodes were wired to picoammeter and LabView was used to acquire the displacement, force and induced charge data. The induced charge during the pull test was measured by the picoammeter. Strain rate was designated at 0.005 mm/s and the test temperature and humidity conditions were 24 °C and 20–25 %RH. Once the induced charge is measured, in-plane piezoelectric charge constant [d31] can be found as [31],

\[ d_{31} = \left( \frac{\partial D_3}{\partial T_1} \right)_E \text{[C/N]} \]  

where, D3 is induced charge per unit electrode area and T1 is applied in-plane normal stress.

2.5. Actuator performance test

Both surfaces of the M/C composites were coated with very thin gold electrodes (150 nm) by using a thermal evaporator.
The size of the samples was $1.2 \text{ cm (width)} \times 5 \text{ cm (length)}$ and the thickness was ranging from $20$ to $23 \mu m$ depending on the stretching ratio. Fig. 2 shows the schematic of aligned M/C composite actuator. The actuator performance was evaluated by measuring bending displacement and resonance frequency. The performance measurement system consists of laser displacement sensor (Keyence, LK-G85 and LK-G15), environmental chamber (KMS, CTH3-2S), current probe (Tektronix, TCP 300), LabView software with personal computer and function generator (Agilent, 33220A) [25]. The prepared sample is placed in the environmental chamber that can control temperature and humidity. The sample is held in a gripper that has electric contacts wired to the function generator and current probe. When the function generator sends out the electrical signal to the sample, the laser displacement sensor measures the bending displacement at the tip of the actuator.

3. Results and discussion

3.1. Chemical, mechanical and electrical characteristics

MWCNTs were functionalized with carboxyl group by nitric acid treatment, and the carboxyl groups were converted into imidazolide by sonochemical reaction with CDI. To investigate possible change in dimension of MWCNTs that can be occurred during functionalization reaction, surface morphology of both functionalized MWCNTs was observed by transmission electron microscopy (TEM) (Fig. 3). Table 1 shows the average length and diameter of the MWCNTs, which are determined from TEM image. The TEM results of the MWCNTs clearly show that not only the length and diameter of MWCNTs are significantly decreased in response to acid treatment, but also further decrease in the dimension can be realized by sonochemical reaction. In contribution of a mild sonochemical process under a low power, the latter can be less significant than a tendency reported [33].

To quantitatively analyze chemical composition of the functionalized MWCNTs, element analysis was performed and the results were correctly fitted by considering an assumption that water molecules can be absorbed on the product due to hydrophilic characteristic of carboxyl groups on MWCNTs. The results were consistent with the following formula of the products: (i) MWCNTs functionalized with carboxyl group: $C\text{–}(\text{COOH})_{0.095}(\text{H}_2\text{O})_{0.05}$ [Calculated (found): %C, 75.9 (76.5); %H, 1.15 (1.14); %O, 22.95 (22.36)] and (ii) MWCNTs functionalized with imidazolide: $C\text{–}(\text{C}_4\text{H}_3\text{N}_2\text{O})_{0.07}(\text{H}_2\text{O})_{0.1}$ [Calculated (found): %C, 75.1 (75.6); %H, 2.0 (2.63); %O, 13.4 (15.9); %N, 9.6 (9.68)]. The percentages of functionalized groups (carboxyl and imidazolide) on MWCNTs with respect to carbon atoms in MWCNTs which are estimated by abovementioned formulas were 25.86% and 32.5%, respectively. These results suggest successful modification of MWCNTs with abundant amount of functional groups (>25%).

By reaction of the imidazolide–MWCNTs with cellulose, MWCNTs were covalently grafted to cellulose. The presence of covalent grafting was investigated by taking evidential FT-IR and Raman spectra, and quantity of the MWCNTs grafted on cellulose was estimated by TGA analysis. In FT-IR spectra of Fig. 4A, the peak at $1375 \text{ cm}^{-1}$ corresponds to the
C–O–H bond of the cellulose [34]. Due to the nitric acid treatment and sonication assisted reaction with CDI, MWCNTs show a carbonyl absorption peak at 1740 cm$^{-1}$ and C–N stretching peak at 1360 cm$^{-1}$, respectively [35,36]. However, after the reaction of MWCNTs with cellulose, the new ester absorption peak was appeared at 1728 cm$^{-1}$. It clearly indicates the presence of covalent grafting of MWCNTs to cellulose chains. In Raman spectra of Fig. 4B, pristine MWCNTs shows D-mode (1290 cm$^{-1}$) and G-mode (1590 cm$^{-1}$) of MWCNTs corresponding to disorder induced mode and tangential mode of MWCNTs, respectively [37]. On the other hand, both functionalized MWCNTs and M/C composite show D mode (1610 cm$^{-1}$) as a shoulder peak of G-mode, which is also associated with defects and disorder induced features in MWCNTs. The remarkable enhancement of D’ mode indicates a new covalent bond formation in the M/C composite.

To estimate quantity of MWCNTs grafted on cellulose, TGA analysis for MWCNTs, functionalized MWCNTs, regenerated cellulose and M/C composite was carried out (Fig. 5). For pristine MWCNTs, there was almost no weight loss (<1%) below 600 °C, and the thermal degradation, which is caused by the presence of disordered and amorphous carbon on the MWCNTs was begun over 600 °C [38]. On the other hand, MWCNTs functionalized with carboxyl groups and imidazole show their weight losses of about 15% and 28% between 100 and 600 °C due to the decomposition of carboxyl and imidazole groups on the surface of MWCNTs, respectively. Especially, for regenerated cellulose and M/C composite, weight loss curve was composed of two stages. The minor weight loss (<5%) at the temperature below 150 °C and the major weight loss at the temperature ranging from 290 to 520 °C was occurred due to evaporation of physically absorbed water molecules and decomposition of cellulose, respectively [39]. In contribution of MWCNTs grafted to cellulose, the temperature for decomposition of cellulose was 10% increased. It indicates that M/C composite exhibits improved thermal stability comparing with regenerated cellulose. By comparing the weight loss of the M/C composite with pristine MWCNTs and regenerated cellulose, the quantity of MWCNTs grafted on cellulose in M/C composite was estimated as 1.35 wt%.

The covalently grafted MWCNTs have improved the mechanical property of cellulose due to their homogeneous distribution in the M/C composite [26]. To further improve the mechanical property of the M/C composite, alignment of MWCNTs in the M/C composite was made by mechanical stretching. Considering that the alignment can be differentiated along with stretching ratio ($S/R$), which defined as a length ratio after and before stretching, the stretching effect was investigated by applying different stretching ratios, namely 1.0, 1.4, 1.6, 1.8 to the M/C composites. Table 2 shows the dependence of stretching ratio on mechanical and electrical properties of aligned M/C composites at 25 °C, 35% RH.

<table>
<thead>
<tr>
<th>Stretching ratio ($S/R$)</th>
<th>Thickness (µm)</th>
<th>Young’s modulus (GPa)</th>
<th>Electrical resistance per unit volume (kΩ/µm$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.0</td>
<td>15</td>
<td>11.4 ± 0.1</td>
<td>113.2</td>
</tr>
<tr>
<td>1.4</td>
<td>13</td>
<td>17.6 ± 0.1</td>
<td>5.23</td>
</tr>
<tr>
<td>1.6</td>
<td>10</td>
<td>20.2 ± 0.2</td>
<td>1.762</td>
</tr>
<tr>
<td>1.8</td>
<td>9</td>
<td>23.4 ± 0.2</td>
<td>0.24</td>
</tr>
</tbody>
</table>
mechanical and electrical properties of aligned M/C composite depending on the stretching ratio. As increasing the stretching ratio up to 1.8, the MWCNTs were more aligned with the cellulose chains, so as to not only enhance its Young’s modulus, but also reduce the electrical resistance (Fig. 6). Since the stretching reduced the space between MWCNTs, the electrical resistance was remarkably decreased. To observe morphological change induced by the alignment of MWCNTs, SEM images were taken at surface and cross-section of the composite. When $S_R$ was reached to 1.8, well aligned bundles of MWCNTs and chains were observed not only on the surface (Fig. 7B), but also in the cross-section (Fig. 7D). The morphology was clearly different with that of the un-stretched composite ($S_R = 1.0$) shown in Fig. 7A and C.

**Fig. 6** – Dependence of stretching ratio on electrical resistance of aligned M/C composites.

**Fig. 7** – SEM images of aligned M/C composites taken at surface ($S_R = 1.0$) (A) and surface ($S_R = 1.8$) (B), cross-section ($S_R = 1.0$) (C) and cross-section ($S_R = 1.8$) (D).

**Fig. 8** – Dependence of humidity on stress–strain curves of aligned M/C composites: $S_R = 1.0$ (A) and $S_R = 1.8$ (B).
On the other hand, the mechanical property of the aligned M/C composites was significantly decreased with increasing humidity level regardless of the stretching ratio. Fig. 8 shows stress–strain curves of the aligned M/C composites depending on the stretching ratio and humidity level. As increasing the humidity level, the yield strength was decreased. The consistent trend was strongly associated with the presence of hydrophilic hydroxyl and carboxyl groups in the M/C composite. In humid condition, the hydrophilic functional groups can easily absorb water molecules from the environment so as to weaken strong hydrogen bonds network between cellulose chains. Furthermore, the amounts of absorbed water molecules that can be changed depending on the humidity level increase the weight and thickness of the M/C composites. Figs. 9 and 10 show the thickness and weight increase of the M/C composite with respect to the humidity level, from 30 to 90 %RH. Significant changes of the weight and thickness predominantly influence in the actuator performance of the aligned M/C composite.

Fig. 9 – Dependence of humidity on thickness of aligned M/C composites.

Fig. 10 – Dependence of humidity on mass of aligned M/C composites.

Fig. 11 – TSC measurement: dependence of stretching ratio on depolarized current of aligned M/C composites.

Fig. 12 – TSC measurement: dependence of poling field on depolarized current of aligned M/C composites.

Fig. 13 – TSC measurement: relationship between maximum depolarized current of aligned M/C composite and poling field.
3.2. Piezoelectric properties

The effect of alignment on enhancing dipolar polarization in aligned M/C composite was analyzed by TSC measurement. Fig. 11 shows the resultant depolarized current profiles of the aligned M/C composites with different $S_R$. The maximum depolarized current was remarkably enhanced by increasing the $S_R$ up to 1.8. Fig. 12 shows the depolarized current profiles of the composites depending on the poling electrical field. As increasing the poling electrical field from 500 kV/m to 1.5 MV/m for the aligned M/C composite ($S_R = 1.8$), the depolarized current was almost linearly increased (Fig. 13). This linear relationship is strong evidence that the aligned M/C composite has piezoelectricity. The results indicate that strong dipolar alignment of MWCNTs and cellulose chains was made by mechanical stretching, which predominantly contributed to enhance its piezoelectric response of M/C composite.

Based on the evidential piezoelectric behavior of the aligned M/C composite proved by TSC measurement, piezoelectric constant $d_{31}$ of the composite was measured with respect to $S_R$ and the orientation direction. Orientation direction is defined as the angle of the aligned M/C composite taken with respect to the stretching direction. For example, $0^\circ$ orientation is the stretching direction. As shown in Table 3, the aligned M/C composite exhibited higher piezoelectric charge constant than that of the cellulose EAPap. Interestingly, $45^\circ$ orientation showed the best $d_{31}$ compared to other orientation in the aligned M/C composite when the $S_R$ is above 1.6. This might be due to the orientation of dipoles, which was established by aligning MWCNTs and cellulose chains. Considering that the highest $d_{31}$ of mechanically aligned cellulose EAPap was found from the $45^\circ$ orientation, the high $d_{31}$ observed from the aligned M/C composite with $45^\circ$ orientation can be strongly associated with not only dipolar orientation of MWCNTs, but also shear piezoelectricity of cellulose [3,4,40].

Table 3 – Dependence of orientation and stretching ratio on piezoelectricity of cellulose EAPap and aligned M/C composites including weight percent of 0.015.

<table>
<thead>
<tr>
<th>Samples</th>
<th>Piezoelectric charge constant (pC/N)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Orientation</td>
</tr>
<tr>
<td></td>
<td>$0^\circ$</td>
</tr>
<tr>
<td>M/C composite ($S_R = 1.0$)</td>
<td>18.1 ± 0.4</td>
</tr>
<tr>
<td>M/C composite ($S_R = 1.4$)</td>
<td>10.0 ± 0.4</td>
</tr>
<tr>
<td>M/C composite ($S_R = 1.6$)</td>
<td>8.5 ± 0.3</td>
</tr>
<tr>
<td>M/C composite ($S_R = 1.8$)</td>
<td>7.8 ± 0.4</td>
</tr>
<tr>
<td>Cellulose EAPap ($S_R = 1.8$)</td>
<td>4.2 ± 0.7</td>
</tr>
</tbody>
</table>

Fig. 14 – Dependence of stretching ratio and humidity on bending displacement of aligned M/C composite actuators.
Alignment effect on the actuator performance of the aligned M/C composite was investigated by measuring their bending displacement and resonance frequency depending on the stretching ratio and humidity level (Fig. 14 and Table 4). As increasing the $S_R$ up to 1.8, the resonance frequency of the actuator and small decrease of its bending displacement. As the humidity level increased from 60 to 90 %RH, the resonance frequency slightly down with the increased bending displacement. The results were strongly associated with not only the enhanced actuating capability of the aligned M/C composite, but also considerable change associated with the improved ion mobility and piezoelectric effect depending on its environmental humidity condition. At low humidity condition, piezoelectric effect of the composite is dominant. Thus, the resonance frequency of the actuator is increased with decreased bending displacement. On the other hand, at highly humid condition, ions, such as $\text{Cl}^-$, can move according to the applied electric field.

### 4. Conclusions

The presence of covalent grafting of MWCNTs to cellulose chains was proved by FT-IR, Raman spectra and TGA analysis. The alignment of MWCNTs with cellulose chains in M/C composite was obtained by mechanical stretching process. Dependence of stretching ratios on alignment of MWCNTs was evaluated by observing morphology as well as measuring mechanical, electrical and piezoelectric properties of the aligned M/C composite. As increasing the stretching ratio ($S_R$) from 1.0 to 1.8, MWCNTs and cellulose chains were more aligned so as to increase the Young’s modulus and piezoelectric constant, and to decrease its electrical resistance. Finally, the influence of the alignment on the actuator performance of the aligned M/C composite was investigated in terms of bending displacement and resonance frequency depending on the stretching ratio and humidity level. The piezoelectric effect enhanced by the alignment was dominant for actuation at low humidity condition. Thus, the resonance frequency of the actuator is increased with decreased bending displacement. On the other hand, at highly humid condition, the predominant actuation principle turned into ion migration effect. As humidity level increased from 60 to 90 %RH, the bending displacement was much improved meanwhile the resonance frequency was a bit slightly down. This result may be strongly associated with the improved ion mobility as well as increased flexibility of cellulose chains due to weaken hydrogen bonds between cellulose chains.

### Acknowledgement

This work is performed under the support of Creative Research Initiatives (EAPap Actuator) of NRF/MEST.

### References


学霸图书馆
www.xuebalib.com

本文文献由“学霸图书馆-文献云下载”收集自网络，仅供学习交流使用。

学霸图书馆（www.xuebalib.com）是一个“整合众多图书馆数据库资源，提供一站式文献检索和下载服务”的24小时在线不限IP图书馆。
图书馆致力于便利、促进学习与科研，提供最强文献下载服务。

图书馆导航:
图书馆首页 文献云下载 图书馆入口 外文数据库大全 疑难文献辅助工具