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PAPER

## Conductive carbon nanotube paper by recycling waste paper

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This paper reports on an easy process of fabricating conductive carbon nanotube (CNT) paper by recycling waste paper. In the absence of dispersion agent, we just mixed and ground multi-walled carbon nanotube (MWCNT) and shredded A4 paper together in water. CNT paper was obtained after filtering and rolling the mixture of MWCNT and waste paper. The thickness of CNT paper was controllable by using the different volume of the mixture in the fabrication. Scanning electron microscope images showed that MWCNT was evenly stuck to the surface of cellulose. Raman spectra indicated that the structure of a sidewall of MWCNT was influenced by cellulose through the mixing process. In addition, thermal stability of CNT paper was enhanced in proportion to the amount of MWCNT. The presence of MWCNT also contributed to the enhancement in the elastic properties of CNT paper. Depending on the amount of MWCNT and the thickness of CNT paper, the sheet resistance of CNT paper varied from 49.1 to 3365.6  $\Omega/\text{sq}$ . This CNT paper can be utilized in the fields of electronic device and energy storage device, leading to easy mass-production at low cost.

### Introduction

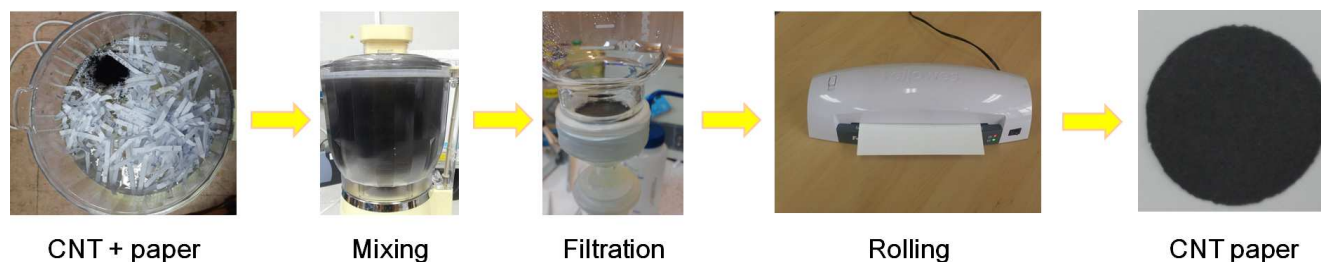
For more than 4000 years, paper has been mainly used for the purpose of recording information. With the advance in the fields of flexible display and wearable device, paper is recently investigated as an alternative to substitute plastic substrates such as polyethylene terephthalate,<sup>1-3</sup> polyethylene naphthalate,<sup>4-6</sup> and polyimide.<sup>7-9</sup> Furthermore, paper becomes more useful in the application to electronics when it has electrical conductivity. And, there have been efforts to fabricate paper having electrical conductivity.

The fabrication method of conductive paper can be classified into two ways. One is to make a composite of a conductive material and cellulose fiber by using a typical process of paper fabrication.<sup>10-15</sup> As a filler to make paper conductive, carbon black,<sup>10</sup> carbon fiber,<sup>10,11</sup> graphite, and carbon nanotube (CNT)<sup>12-15</sup> were adapted recently. The other is to coat paper with conductive materials by using conductive ink<sup>16-17</sup> as well as conductive polymer.<sup>18</sup> In both methods, CNT is utilized as a material to make paper electrically conductive because CNT has superior electrical conductivity. Hu et al. fabricated a highly conductive paper by coating CNT ink on paper.<sup>16</sup> The sheet resistance of the conductive paper was controlled in the range from  $10^5$  to  $10$   $\Omega/\text{sq}$  by varying the thickness of paper. As a modified method, cellulose sheet was dipped in a solution where CNT is dispersed evenly, and conductive paper was successfully obtained.<sup>19</sup>

In spite of using CNT as superior conducting filler, there is

still a technical barrier to be improved. In preparing conductive paper by the method of mixing CNT and cellulose, we should conduct a typical procedure of obtaining cellulose fiber by chemical modification of pulp, which is a cost and time consuming process. In addition, the use of a surfactant is inevitable to achieve good dispersion of CNT solution. Meanwhile, coating CNT on paper is relatively easier than the typical method mentioned above, but we cannot avoid preparing an ink containing conductive material. This is another kind of a laborious task. In this case, the use of a surfactant cannot be also avoided in the step of conductive ink formulation when CNT is involved. In any case, the process to remove the surfactant is required to make paper conductive. Detailed reason can be explained as following: generally, paper becomes electrically conductive due to CNT which is continuously connected as a structure of networks on paper. We can expect electrical conductivity from the percolated CNT networks constituting electrical paths in paper. However, CNT networks do not play a role as an electrical path in the presence of a surfactant since CNT is covered with the surfactant which is an insulator electrically. Therefore, post-process is required to remove the surfactant from conductive paper in all cases.<sup>20</sup> Furthermore, the chemical procedure of obtaining cellulose from pulp is preferably avoided in the fabrication of conductive paper if possible.

To overcome this problem, we show a simple way of preparing conductive paper by using CNT and waste paper without a surfactant. To evade the chemical treatment to obtain cellulose fiber, we used waste paper (shredded A4 paper) as a source of ce-



**Fig. 1.** Simple procedure of preparing conductive paper by using MWCNT and shredded A4 paper. Conductive paper is easily prepared by mixing MWCNT and shredded A4 paper followed by the processes of filtration and rolling. The rolling was conducted by covering CNT paper with two sheets of paper.

llulose for the fabrication of conductive paper. This method is attractive in the respect of recycling waste paper as well as avoiding a chemical treatment occurred in the fabrication of paper. And, multi-walled carbon nanotube (MWCNT) was employed as a material to make paper electrically conductive. Our idea is to mechanically grind and mix shredded paper and MWCNT together in water to obtain the slurry of cellulose and MWCNT. Through the mechanical mixing, we can expect good dispersion of MWCNT in the slurry without using a surfactant. After the mixing, conductive paper is achieved *via* filtering and rolling the slurry. By varying the amount of MWCNT and the volume of the slurry, the electrical conductivity and thickness of conductive paper was controllable.

Apart from mass production, our method is time- and cost-effective compared with the mentioned methods on fabricating conductive paper. We think that CNT paper can be applied to diverse fields of electrode of energy storage device, flame retard, and electromagnetic shielding as well as flexible substrate.

## Experimental

### Preparation of CNT paper

Fig. 1 shows the process of fabricating conductive paper by recycling waste paper and CNT. As shown in Fig. 1, we used shredded A4 paper as a source of cellulose and MWCNT (Nanocyl NC 7000, average diameter: 9.5 nm, average length: 1.5  $\mu\text{m}$ , purity: 90%, surface area: 250 - 300  $\text{m}^2/\text{g}$ ) as a conductive material. A kitchen mixer grinder was used to obtain the slurry of MWCNT and cellulose as shown in the mixing process of Fig. 1. MWCNT and 1.5 g of shredded paper were added in 500 ml of de-ionized water and mixed for 4 min at 15,000 rpm. To control the electrical conductivity of paper, a different amount of MWCNT (3, 7, and 10% wt.) was added in the preparation of the slurry. After mixing, the slurry was filtered using a filter paper (ADVANTEC, 5C, diameter: 5.5 cm) by vacuum and washed with 100 ml of de-ionized water to remove the unknown chemicals from waste paper. In this step, various volumes of the slurry (10, 15, and 20 ml) were used to fabricate paper having different thickness. Next, the filtered slurry was dried at 120  $^{\circ}\text{C}$  and pressed by office laminator to obtain paper having even and smooth surface. As a reference sample, we prepared paper by using 20 ml of the slurry without MWCNT (hereafter, reference).

### Characterization

Scanning electron microscope (SEM, MIRA 3 FEG SEM) was

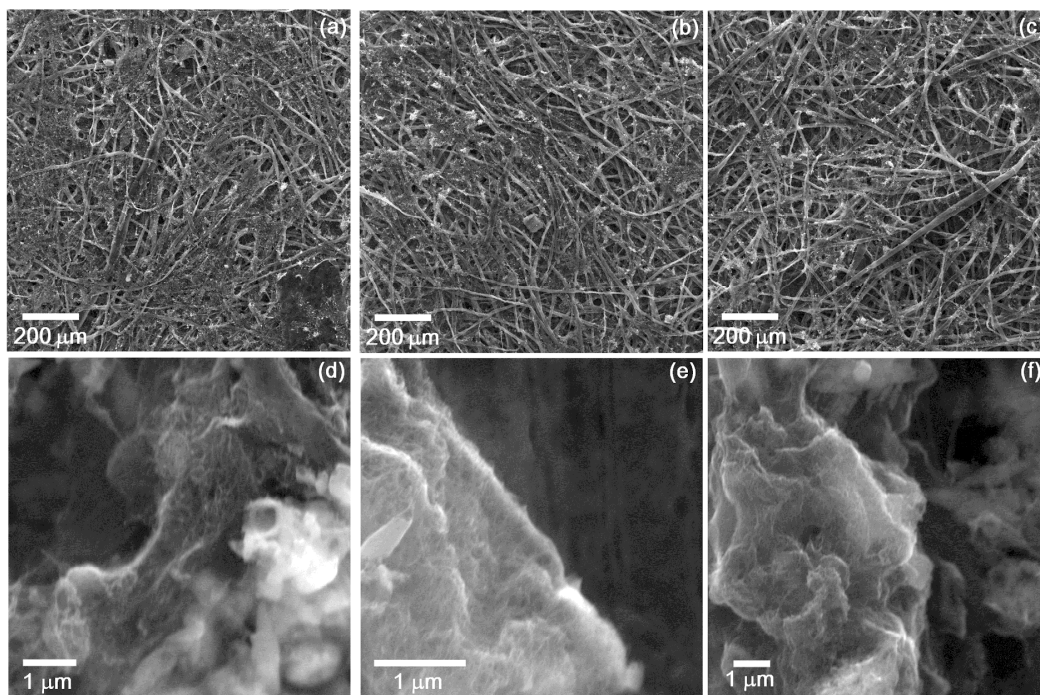
employed to observe the surface of CNT paper and the morphology between MWCNT and cellulose. The presence of MWCNT in the paper was confirmed by Raman spectroscopy (HORIBA Jobin-Yvon, HR-800UV, Ar laser, 514.5 nm). To investigate the thermal stability of CNT paper, thermogravimetric analysis (TGA, TA Instruments, Q500) was conducted. Temperature was raised up to 450  $^{\circ}\text{C}$  at a 10  $^{\circ}\text{C}/\text{min}$  ramp rate. By using 4-point probe method (Advanced Instrument Technology, CMT-series), sheet resistance of CNT paper was measured at six different sites of each paper and the average value was used as sheet resistance of the CNT paper. By using digital thickness gage (Mitutoyo Corp.), the thickness of CNT paper was measured in the same way as performed in measuring sheet resistance. To show that CNT paper is reproducible, three sheets of CNT paper were used in the analysis of sheet resistance and thickness. To study the elastic properties of CNT paper, Young's modulus was measured with a universal test machine (Instron 5582) at a cross-head speed of 1 mm/min.

CNT papers which were prepared by using 20 ml of the slurry at the concentration of 3, 7, and 10% wt. were used in the analysis for SEM, Raman spectroscopy, and TGA (hereafter, 3, 7, and 10%). For the measurement of Young's modulus, 200 ml of the slurry at the concentration of 3, 7, and 10% wt. were filtered by using Buchner funnel with a filter paper (CHMLAB GROUP, No, F2040-110, diameter: 11 cm) to obtain a large size of CNT paper.

## Results and discussion

We conducted SEM analysis to observe the morphology of cellulose and MWCNT in CNT paper. Fig. 2 represents the SEM images of 3, 7, and 10% prepared by using 20 ml of the slurry. The observation was conducted at low (Fig. 2 a, b, and c) and high (Fig. 2 d, e, and f) magnification, respectively. The images seen in Fig. 2 a, b, and c present the morphology of cellulose fibers observed in the surface of each paper. We can see from the image that cellulose fibers are tangled together in all cases, meaning that our method of grinding and filtration was successful in fabricating paper. Despite we used shredded paper as a cellulose source, the morphology of a tangled structure is consistent with that of other works on paper fabrication.<sup>10-13,16-19,21</sup> In order to see the morphology between cellulose and MWCNT, we observed CNT paper at high magnification. Fig. 2 d, e, and f represent the images obtained 3, 7, and 10% CNT paper, respectively. From Fig. 2d, we can notice that MWCNT



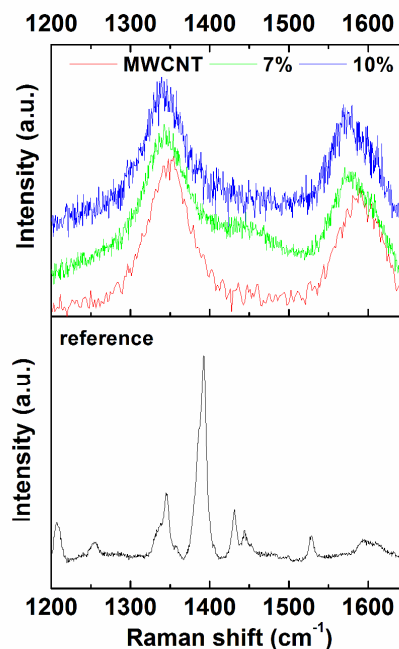


**Fig. 2.** SEM images of CNT paper containing 3, 7, and 10% wt. of MWCNT at low (a, b, and c) and high (d, e, and f) magnification, respectively. 20 ml of the slurry was used in the preparation of each paper. A tangled structure of cellulose fiber is observed from all CNT papers, and MWCNT covers the surface of cellulose fiber in all cases. This means the mechanical mixing was significant to good dispersion of MWCNT on the surface of the cellulose, substituting the usage of dispersion agent for MWCNT.

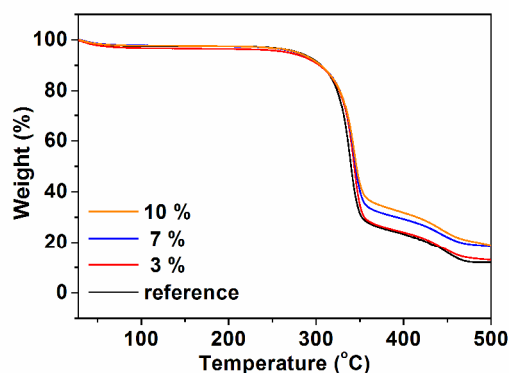
exists in the surface of cellulose even in a small amount. When the weight percentage of MWCNT increased up to 7 and 10% in CNT paper, more MWCNT was observed in the surface of cellulose as seen in Fig. 2 e and f. Interestingly, the SEM images confirm that MWCNT is evenly dispersed on the surface of cellulose without using dispersion agent in the process of the mixing. This means that the mechanical mixing of shredded paper and MWCNT in water is significant to good dispersion of MWCNT in the surface of cellulose. In other words, the mechanical mixing can substitute the usage of dispersion agent for MWCNT in the preparation of conductive paper. We think that a morphological similarity between MWCNT and cellulose also contributed to a well-woven structure of CNT paper.

Raman spectroscopy is a useful way to detect the existence of MWCNT because it is sensitive in the Raman shift range around 1350 and 1590  $\text{cm}^{-1}$ . Furthermore, the effect of cellulose on MWCNT can be analysed by inspecting a change of spectra in the ranges. Therefore, we conducted Raman spectroscopic analysis on CNT paper to clarify the presence of MWCNT and the mutual influence between cellulose and MWCNT. Fig. 3 exhibits the Raman spectra measured from reference, 3, 7, and 10%. To confirm the Raman spectra originated from cellulose, we obtained the spectra from reference paper. The result was displayed in the bottom of Fig. 3. As shown in the figure, several spectra are detected from the reference paper and the strongest intensity appears at 1392  $\text{cm}^{-1}$ . Then, the Raman spectra of MWCNT were measured as displayed in the top of Fig. 3. We can see two major spectra at 1374 and 1591  $\text{cm}^{-1}$ . The spectra at 1374  $\text{cm}^{-1}$  has been known as D-band which is related to defects or

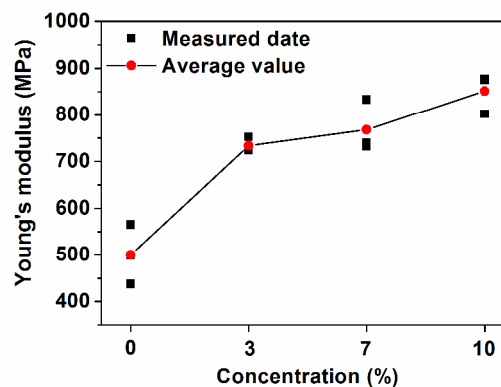
disorder in CNT. Meanwhile, G-band, originated from  $\text{sp}^2$ -hybridized



**Fig. 3.** Raman spectra of reference (black), MWCNT (red), 7% (green), and 10% (blue) prepared by using 20 ml of the slurry. In the case of 3%, the spectra corresponding to MWCNT were not detected. The spectra measured from CNT paper were shifted to the left, compared with the original spectra of MWCNT. This means that the structure of a sidewall of MWCNT was modified by mixing with cellulose.



**Fig. 4.** TGA analysis of reference (black), 3% (red), 7% (blue), and 10% (orange) prepared by 20 ml of the slurry. The thermal stability of CNT paper is prominently enhanced when the concentration of MWCNT reaches 7%.



**Fig. 5.** Young's modulus of reference, 3%, 7%, and 10%. Black square indicates the measured data from three sheets of paper while red circle exhibits the average Young's modulus at each condition. The presence of CNT in paper enhances the Young's modulus of paper in proportion to the concentration,

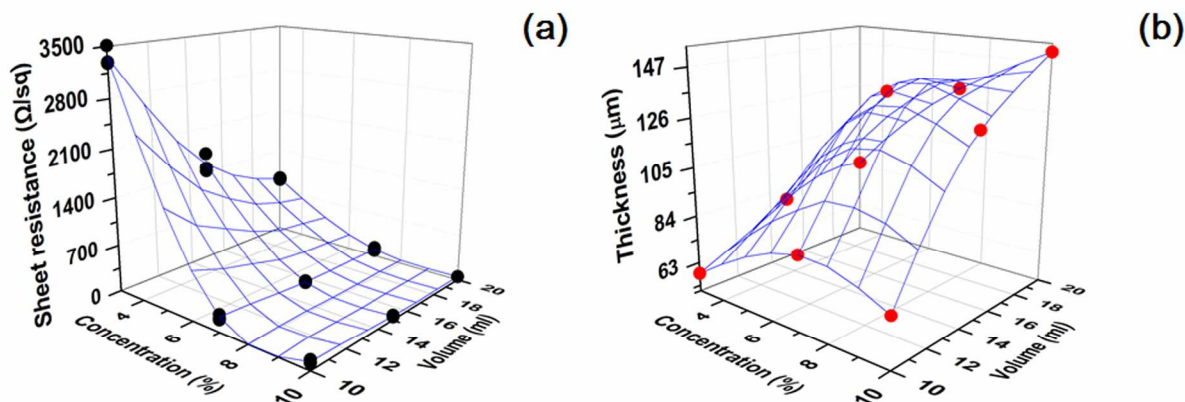
dized carbon in honeycomb structure, has been reported to appear around  $1590\text{ cm}^{-1}$ . In the case of 3%, we detected only the spectra of cellulose than the ones corresponding to MWCNT even though the paper showed a gray colour. This means that 3 wt. % is not the concentration enough to be detected in Raman spectroscopy analysis although we observed MWCNT in SEM analysis. In the case of 7%, we can see that D- and G-band spectra appear at  $1343$  and  $1570\text{ cm}^{-1}$ , respectively. And, no spectra are observed around  $1392\text{ cm}^{-1}$  where the reference paper shows the strongest intensity in the measured Raman spectra. In addition, the presence of cellulose shifted D- and G-band spectra to the left, compared with the spectra measured from MWCNT. The same phenomenon occurs in the case of 10%. Both D- and G-band are shifted to the left and positioned at  $1339$  and  $1570\text{ cm}^{-1}$ , respectively. According to the review on Raman spectra of CNT by Dresselhaus and co-workers,<sup>22</sup> the shift of G-band spectra breaks out when structural modification occurs in a sidewall of MWCNT by the attachment of other chemical species. Therefore, our result indicates a possibility that the structure of a sidewall of MWCNT has been modified by mixing MWCNT with cellulose together. Also, this is supported by our SEM analysis showing that MWCNT was attached to the surface of cellulose.

MWCNT has been known to show superior properties thermally and mechanically. Hence, the presence of MWCNT in paper can enhance its thermal stability, depending on the concentration of MWCNT. Therefore, TGA analysis was conducted to investigate the thermal stability of CNT paper by the amount of MWCNT. Fig. 4 gives weight loss with respect to temperature of reference, 3, 7, and 10%, respectively. The previous study reported that paper slightly lost a weight below  $100\text{ }^{\circ}\text{C}$  due to water and that a weight loss by the thermal decomposition of cellulose occurred drastically above  $300\text{ }^{\circ}\text{C}$ .<sup>23</sup> The same phenomenon appears in Fig. 4, irrespective of the presence of MWCNT. And, all papers show a steep decrease in weight loss when temperature is higher than  $300\text{ }^{\circ}\text{C}$ . Then, the thermal decomposition proceeds slowly as the temperature reaches to  $350\text{ }^{\circ}\text{C}$ . This result is also consistent with the result of the work mentioned above. However, we can see that all papers have a different decomposition rate at  $350\text{ }^{\circ}\text{C}$ , depending on the

amount of MWCNT in CNT paper. Reference and 3% show a similar behaviour in weight loss above  $350\text{ }^{\circ}\text{C}$ . Meanwhile, 7 and 10% result in a less weight loss at  $350\text{ }^{\circ}\text{C}$ , meaning that the increased content of MWCNT contributed to the enhancement of thermal stability of CNT paper. We can notice that 10% exhibits the strongest thermal stability among the prepared CNT papers. From this, we can deduce that the thermal stability of CNT paper can be enhanced significantly when the concentration of MWCNT is at least 7%.

To see the effect of the presence of CNT on the elastic properties of CNT paper, we measured Young's modulus of each CNT paper as well as reference. Three sheets of paper were used for the measurement, and the results were plotted in Fig. 5. The figure displays the average Young's modulus (red circle) of measured data with raw data (black square). As shown in Fig. 5, Reference has the average value of  $500.4\text{ MPa}$  and Young's modulus drastically increased up to  $733.8\text{ MPa}$  with the addition of MWCNT. When the concentration of MWCNT is 7%, Young's modulus of CNT paper was enhanced from  $733.8$  to  $768.0\text{ MPa}$ . And, the maximum value of  $851.2\text{ MPa}$  was obtained as the concentration increased up to 10%. This means that the presence of CNT enhances the elastic properties of CNT paper in proportion to the concentration.

By varying the volume of the slurry and the amount of MWCNT, CNT paper can have different thickness and electrical conductivity. To clarify the influence of the MWCNT concentration and thickness on electrical conductivity, we prepared conductive papers which have different thickness and electrical conductivity, and measured the thickness and sheet resistance of CNT papers. Fig. 6a represents a plot of the sheet resistance of conductive paper versus the concentration of MWCNT and the volume of the slurry. As shown in the figure, we can notice that sheet resistance gradually decreases as the concentration increases in CNT paper. Sheet resistance is varying from  $49.1$  to  $3365.6\text{ }\Omega/\text{sq}$ , depending on the amount of MWCNT. This means that the conductivity of paper can be controlled by varying the concentration of MWCNT. This also points out that MWCNT are networked quite well in the paper, providing an electrical path for electron. Fig. 6a shows the effect of volume of the slurry on sheet resistance. When the concentration is fixed to

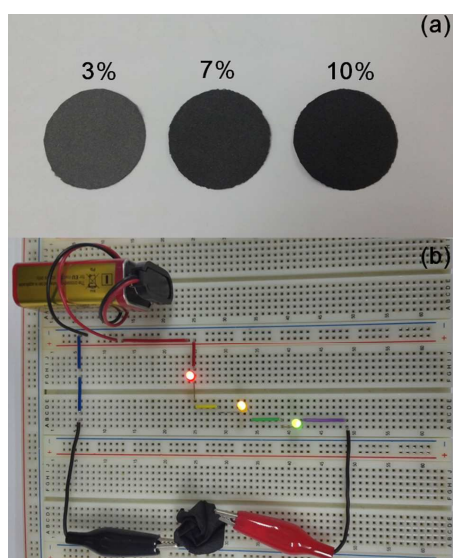


**Fig. 6.** Surface plot of (a) sheet resistance and (b) thickness of CNT paper with respect to the MWCNT concentration and the volume of the slurry. The surface was plotted by using the average value of sheet resistance and thickness obtained from three sheets of paper. Black circle in (a) indicates the measured sheet resistance at each condition while red circle in (b) displays the average thickness. The increase in the concentration and the volume decreases the sheet resistance of CNT paper. Meanwhile, the thickness of CNT paper is only a function of the volume of the slurry.

3% wt., the sheet resistance of CNT paper drastically decreases with the increase in the slurry volume. Meanwhile, the trend of a sudden decrease in sheet resistance does not appear at the concentration of 7 and 10% in spite of the increased volume of the slurry. This means that 7 and 10% are the proper concentration constituting electrical path in paper at the small volume of the slurry, leading to low sheet resistance. On the other hand, 3% is the concentration influenced by the volume of the slurry because sheet resistance decreases with respect to the increase of the volume. That is, we cannot expect enough electrical paths by CNT network when the paper is prepared by 10 ml of the slurry at 3 wt. %. However, the increase of the slurry volume provides more electrical paths as a form of CNT networks in the paper, resulting in the decrease of sheet resistance from 3365.6 to 895.5  $\Omega/\text{sq}$ . Fig. 6b presents a change of the thick-

ness with respect to the CNT concentration and the slurry volume. The figure reveals that regardless of CNT concentration, the thickness of CNT paper becomes thick in proportion to the volume of the slurry. The maximum and minimum in the thickness of paper are 66.7 and 107  $\mu\text{m}$ , respectively. From Fig. 6b, we can notice that the concentration of CNT contained in paper do not influence the thickness of paper.

We folded origami rose to show the CNT paper is foldable without losing electrical conductivity. Fig. 7a indicates the CNT papers prepared with different concentrations of MWCNT based on shredded paper. The image reveals that the colour of paper becomes close to black with the increase of MWCNT concentration. In all cases, MWCNT was not stuck to hands when we treated the CNT papers, meaning that MWCNT was captured well whining paper as seen in SEM images. However, when the concentration increased to 15% wt., MWCNT was stuck to hands in touching and folding CNT paper. This is the reason why we employed 10% wt. of MWCNT in the fabrication of CNT paper. As mentioned before, the thickness of CNT paper can be controlled by changing the volume of the slurry. This means that we can easily prepare well-foldable CNT paper which keeps good electrical conductivity. Fig. 7b displays origami rose prepared by CNT paper, which plays a role as a part of circuit. This indicates that foldable and conductive paper can be prepared compatibly.



**Fig. 7.** (a) CNT paper prepared by using 20 ml of the slurry and (b) origami rose connected to the circuit. CNT paper is foldable and conductive compatibly as shown in the figure.

## Conclusions

We successfully demonstrated the method to fabricate conductive CNT paper by grinding and mixing shredded paper and MWCNT in water. Without using a surfactant, the mechanical grinding led to even dispersion of MWCNT on the surface of cellulose. The presence of MWCNT in paper enhanced the thermal stability of CNT paper and shifted the G-band spectra of Raman spectroscopy to the left in the Raman shift. The sheet resistance and thickness of CNT paper were controllable by varying the amount of MWCNT and the volume of the slurry in the step of the paper fabrication. The maximum and minimum sheet resistances were 3365.6 and 49.1  $\Omega/\text{sq}$ , respectively. In addition,



the thickness of CNT paper was controlled from 66.7 to 107  $\mu\text{m}$ .

We think that this work will open a potential avenue to mass production of conductive paper as well as recycle of waste resource.

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## 10 Notes and references

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