1 Introduction
During the past century, electronic systems have undergone innovative development with assembly and fabrication technology. Architecture of printed circuit boards and photolithography allow for miniaturization and high performance of electronic systems. Present electronic devices are mostly manufactured using rigid or flexible printed circuit boards. However, future electronic applications such as wearable computers [1], conformable skin sensors [2] and personal health monitors [3] require stretchable systems to enhance the comfort of the user. The electronic circuits in these applications need not only flexible but also stretchable. Therefore, stretchable interconnection technology has attracted increasingly significant interest to realize high conductive circuits with large deformations.
In the past years, a few research groups [4-8] have reported their efforts on the development of stretchable electronic circuits. One of the approaches is metallic patterning on or in stretchable substrates with an optimized in-plane circuit design; although it is known that free-standing metallic thin films rupture under a tensile strain of ~1 or 2% [9], meandering structures of metallic interconnection lead to stretchability [10]. This approach is advantageous for easy application to the current fabrication technology by replacing rigid or flexible substrates with stretchable elastomers. On the other hand, this is restricted to specific expensive electronic applications and degree of freedom for design of metallic materials, for example composites or alloys. Because most of previous researches using this concept utilize conventional fabrication methods which are based on complicated photolithography technology or vacuum processes.

Direct printing technology such as inkjet, screen, and gravure printing is considered as a promising alternative to the conventional fabrication methods due to the advantages of an additive manufacturing process videlicet low cost, reduction of waste materials and no toxic chemical etchants [11-15]. Especially, screen printing allows for the low-cost fabrication and ease of mass production, owing to its relatively high throughput, simple equipment and usability of a wide range of nanopastes [16, 17]. The conductive and robust fillers of nanopastes are required for fabrication of stretchable interconnections by screen printing, due to weak bonding strength and voids between metallic nanoparticles. The combination of metallic nanoparticles and multi-walled carbon nanotubes (MWNTs) is favorable for highly conductive and mechanically durable fillers [18].

Our strategy to obtain stretchable circuits is development of composite nanopaste using silver (Ag) nanoparticles and MWNTs, and fabrication by screen printing with various circuit geometries. The mechanical properties of the screen-printed Ag-MWNT circuits were evaluated by monotonic and cyclic tensile tests, and the fatigue behaviors under tensile deformations were analyzed by measuring electrical resistance and by observing fractures of circuit surfaces.

2 Experimental Details
2.1 Manufacturing of Composite Nanopaste
The Ag-MWNT composite nanoparticles were produced by the synthesis method suggested by Hong’s group [19-21]. MWNTs were functionalized...
to get stable dispersion in ethanol, and they were homogeneously mixed with a Ag salt. Ag oxide nanocomposite particles were formed on the MWNT surfaces by heating the solution in air. The Ag-MWNT composite nanoparticles were obtained through the calcination and reduction process in a hydrogen atmosphere. Using these composite nanoparticles, the Ag-MWNT nanopaste was finally manufactured by mixing with binder and dispersing agent in an organic matrix based on an α-terpineol solvent. The size of Ag nanoparticles was distributed from 10 nm to 200 nm, and their mean diameter was approximately 50 nm. The Ag-MWNT composite nanopaste had a solid loading of around 60 wt.%, and contained different fractions of the MWNT from 0 to 3 wt.%.

2.2 Circuit Design for Stretchable Interconnection

The stretchable interconnection was designed with three different circuit shapes (rectangular pulse, horseshoe and zigzag), and they were compared with a single-line circuit as a reference. Figure 1 shows the exact dimensions and geometries of each circuit design. The width of all circuits and length of all samples were fixed to 300 µm and 100 mm, respectively. The square pads were positioned at both ends of all circuits, and their size was 5×5 mm².

2.3 Specimen Preparation

A stencil mask of each circuit design was fabricated using a laser drilling method. All circuits were duplicated on an 100-µm-thick thermoplastic polyurethane (TPU) by a screen printing machine (MT-550TV, Micro-Tec, Japan) with a 400-mesh stencil mask. Square patterns were also screen-printed to investigate the effect of MWNT composition ratio on the electrical resistivity. All the printed patterns and circuits were dried on a hotplate at 70 °C for 10 min, and they were then sintered at 140 °C for 1 h in air using a box-type muffle furnace (RTA-BRT100, BLS Korea Inc., Korea). After heat treatment, the printed circuits were encapsulated with another TPU layer. For each experimental condition, twelve samples were prepared for statistical accuracy.

2.4 Characterization of Ag-MWNT Nanocomposite Circuits

High resolution transmission electron microscopy (HRTEM) was employed to evaluate the dispersion states of the Ag-MWNT composite nanoparticles. The microstructure of the Ag-MWNT circuit surface was observed by field emission scanning electron microscopy (FE-SEM). The specific resistivity of the Ag-MWNT films on the TPU substrate was measured by a four-point probe method. Stretchability of the Ag-MWNT circuit was assessed by a monotonic tensile test, and the cyclic endurance reliability was evaluated by a cyclic tensile test. The speed of monotonic tensile testing was 2.5 mm/min. The criterion for extensibility of circuit was determined by the strain required for circuit failure electrically. The cyclic tensile test was performed by stretching the samples at a strain rate of 1% with a frequency of 0.5 Hz, and their electrical resistance

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Fig. 1. Schematics of the various circuit designs: (a) single line, (b) rectangular pulse, (c) horseshoe, and (d) zigzag.
was monitored. While the Ag-MWNT circuits were being stretched, the cracks on the surfaces were observed with optical microscopy to investigate the effects of MWNT ratios and circuit designs on the stretchability of the circuits.

3 Results and Discussion

Figure 2 shows a typical HRTEM micrograph of the Ag-MWNT composite nanoparticles used in this study. This HRTEM micrograph clearly shows that tens nanometer-size Ag nanoparticles were synthesized and most of them were attached on the surface of the MWNTs. The MWNTs were well dispersed without agglomeration, and their diameter ranged between 10 nm and 20 nm. When the MWNTs were distributed in a nanoparticle matrix, the agglomeration between the MWNTs induced by their high Van der Waals force and aspect ratio.

![HRTEM micrograph of Ag-MWNT composite nanoparticles](image)

Fig. 2. A typical HRTEM micrograph of the Ag-MWNT composite nanoparticles.

![FE-SEM micrographs of Ag-MWNT circuit surfaces](image)

Fig. 3. FE-SEM micrographs of the Ag-MWNT circuit surfaces with various durations of sintering and the MWNT ratios (All scale bar is 300 µm.).
could be suppressed [22]. As the circuit contains more MWNTs, they and their networks were observed on the surface before sintering. However, most of them disappeared on the surface after sintering, and they were densely distributed near a few localized surface pores. This is because the MWNTs interrupt coarsening of Ag nanoparticles around MWNTs during sintering [23]. The FE-SEM micrographs depict the microstructural evolution consists with the tendency; the circuit containing the MWNT of 3 wt.% shows the most porous surface. The Ag-MWNT circuits sintered at 140 °C for 1 h show that Ag nanoparticles changed into Ag or Ag-MWNT clusters forming networks via interparticle necking. Though the sintering time was longer than 1 h, the similar microstructure was observed compared with that of the circuit sintered for 1 h, which is in agreement with the result of our previous study [24]. During the sintering process, atomic diffusion drives nanoparticle surface elimination such as evaporation of organic solvent and dispersing agent, leading to the neck formation and destruction of pores between fillers. It is known that the driving force for sintering mechanism is the change in free energy to be minimized the surface free energy [25].

Figure 4 presents the influence of the MWNT ratios on the cluster size and the electrical resistivity of the Ag-MWNT composite nanopaste. Fig. 4 (a) reveals that the cluster size increases with increasing sintering time or the MWNT ratio. As the composition ratio of the MWNT increased from 0 to 3 wt.%, the cluster size of the circuits sintered for 1 h became approximately 1.7 times larger. Though the porous microstructure and MWNT agglomeration were observed on the surface of circuits with higher MWNT ratios, the Ag nanoparticles staying away from MWNTs formed the larger clusters and drove the elimination of pores located in intersections of grain boundaries. Fig. 4 (b) shows the relation between the MWNT ratio and electrical resistivity of the circuits sintered for 1 h at 140 °C. The lowest electrical resistivity of the Ag-MWNT circuit was achieved at the largest amount of 3 wt.%: The measured value was 37.16 µΩ cm, which is around 23 times larger than the electrical resistivity of Ag bulk. It means that the Ag-MWNT composite nanoparticles contribute to the formation of conductive paths and the creation of continuous connectivity electrically despite of the porous microstructure.

Figure 5 shows the stretchability of the Ag-MWNT circuits with respect to the MWNT ratio and circuit geometry. The stretchability of the Ag-MWNT circuits was proportional to the MWNT composition rate for all kinds of circuit geometries. It is observed that the stretchability is dramatically enhanced by the MWNT ratio of 1 wt.% and gradually increased with larger amount of MWNT ratios (> 1 wt.%). Overall, the stretchability of the circuit regardless of the circuit geometry was 2.5-fold increase as the MWNT ratio increased from 0 to 3 wt.%. For the effect of the circuit design, the monotonic tensile test results demonstrated that the stretchability increased
Fig. 5. The stretchability of the Ag-MWNT circuits with various composition rates of MWNT.

in the order of single line < zigzag < rectangular pulse < horseshoe. The maximum of stretchability (~45%) was obtained when the horseshoe-type circuit containing the 3 wt.% MWNT.

After the monotonic tensile test, the fractures of the circuits were observed with optical microscopy, as shown in Fig. 6. The widths of the circuits containing the 3 wt.% MWNTs show further wider than those of the Ag circuit due to the different optimized printing parameters such as a squeeze speed, its angle, and a gap between the stencil mask and the TPU substrate. The viscosity of the Ag-MWNT composite nanopaste (3 wt.% MWNT) was around 10.2 times higher than that of Ag nanopaste (0 wt.% MWNT). This disparity of viscosity leads to the different transfer amount of nanopaste onto a substrate under a same pressure during the squeeze stroke. Small arrows in Fig. 6 are situated at the cracks generated by uniaxial tensile strain. In fact, similar crack distributions and fractures were observed regardless of the MWNT contents. Otherwise, the different path of crack propagation was investigated in comparing the different composition rate of MWNT. It is assumed that the MWNTs play an important role as an obstructor when the cracks propagate between the loosely-packed clusters or non-evolved nanoparticles. Single-line circuits show several thick cracks distributed irregularly on the surface. Compared with the single line, the other geometries show smaller cracks on the apexes or curved edges. These crack distributions mean that the meandering geometries are favorable for reduce of strain accumulation when they deform out of plane [26]. From these fracture analyses, it is concluded that the stretchable interconnect can be achieved by modification of material structures as well as design of circuit geometry.

Figure 7 shows the profile of the electrical behaviors for the Ag-MWNT circuits during cyclic tensile testing. The normalized electrical resistance \((R - R_0)/R_0\) of the Ag-MWNT circuits fluctuated up and down until the variation suddenly reached a critical

Fig. 6. Optical micrographs of the cracked surfaces of the Ag and Ag-MWNT circuits designed with various shapes. (All scale bar is 500 µm.)
Fig. 7. Normalized resistance variation of the Ag-MWNT circuits containing various MWNT ratios during cyclic tensile testing: (a) single line, (b) rectangular pulse, (c) horseshoe, and (d) zigzag.

point, circuit failure. When the circuits were stretched the connections between clusters or particles were easily destroyed or partially necked. Conversely, when the strain on the circuits was removed some relaxation took place due to the stability of TPU substrate. This cyclic deformation of the circuits caused the fluctuation in the electrical resistance. The stretching cycle required for circuit failure increased with increasing the MWNT ratios. The maximum stretching cycle was achieved by the horseshoe circuit containing 3 wt.% MWNT. Regardless of the circuit geometry, the 3 wt.% MWNT circuits show the wide range of fluctuation during cyclic tensile testing. It is indicated that the MWNTs contribute the enhancement of the electrical stability by two possible mechanisms, the suppression of crack propagation and robust connections between conductive fillers. The results of cyclic tensile test show that the cyclic strain resistance is the same tendency with the stretchability measured by the monotonic tensile test.

4 Conclusions

The Ag-MWNT composite nanopaste enables the construction of stretchable and printable circuits on elastomeric substrates. We achieve the stretchable circuits screen-printed on the TPU substrate with various circuit geometries. The stretchability and fatigue resistance were assessed by the monotonic and cyclic tensile tests. The experimental results reveal that significant enhancement is obtained by design of material structures and circuit geometries: the horseshoe-type circuit containing 3 wt.% MWNT presents the largest stretchability and the most robust cyclic resistance. We expect that
feasible application areas can be derived from our novel approaches to stretchable and printable electronic systems.

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References


[22] Y. Hao, Z. Qunfeng, W. Fei, Q. Weizhong and L. Guohua “Agglomerated CNTs synthesized in a fluidized bed reactor: agglomerate structure and


