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Development and Improvement of a "Paper Actuator" Based on Carbon Nanotube Composite Paper with Unique Structures

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Abstract: We propose a new type of soft actuator based on carbon nanotube (CNT) composite paper (CNTCP), i.e., a paper actuator. In our previous study, we demonstrated that actuator operation was possible when using CNTCPs as electrodes with ordinary paper containing ionic liquid between the electrodes; however, their bending motion was not sufficient. Therefore, we here attempt to modify the paper actuator. For this, we tried to soften CNTCPs by first reducing the ratio of contained CNTs. In addition, as a new strategy, we took advantage of the fact that the proposed actuator was made of paper and introduced the *Kirigami* (introducing periodical slits to papers) technique into the structure of our paper actuator. As a result, the performance of the actuator was improved, and its bending motion became visibly larger. The response of the improved actuator to the input voltage was investigated in detail, and the detailed operating conditions could be clarified. Moreover, it was found that not only a bending motion but also a twisting motion could be realized in specific slit patterns. It is thought that the fact that the variation in movement can be increased simply by adding incisions is unique to the proposed paper actuator.

Keywords: carbon nanotube; actuator; paper; carbon nanotube composite paper; ionic liquid; *Kirigami* technique; bending motion; twisting motion

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1. Introduction

In recent years, great progress has been made in nanotechnology research. Among nanotechnology research, studies on nanocarbon materials, such as fullerene, carbon nanotubes (CNTs), and graphene, are being conducted in a wide range of fields, from fabrication techniques to applications. In particular, CNTs, discovered in 1991 [1], are known for their high chemical stability, mechanical strength, high electrical as well as thermal conductivity, and metallic as well as semiconducting electrical properties [2–7]. Because of the various beneficial characteristics described above, there are great expectations for the practical application of various applied articles using CNTs [8]; however, because CNTs are generally a nanoscale material, with a diameter of only a few nm and a length of only a few µm, and because most commercial products on the market are in powder form or a liquid dispersion, they are difficult to handle as they are, making it difficult to develop applications for them. Therefore, research is being conducted not only on methods that use CNTs alone, but also on applying them to devices in the form of composite materials that combine them with other materials, i.e., one solution is to mix CNTs with other materials and handle them as "CNT composite materials". By making a composite material, handling becomes easier, and the functions of CNTs can be used in this form [9–12].

We are developing "CNT composite papers [13]", which can be easily handled as "familiar objects" with the various features of CNTs. These composite materials have attracted attention as unique new materials because they have the same processability and deformability as papers, while maintaining the various functions of CNTs. Various applications of the composites are already under investigation, including the feasibility of "paper transistors [14]", "paper dye-sensitized solar cells [15]", and "thermoelectric power generating papers [16,17]".

Recently, as technology has advanced, there have arisen demands for actuators that are smaller, lighter, and more flexible; therefore, various soft actuators with these characteristics have been developed. In particular, CNTs are attracting attention as one of the materials for these soft actuators. For example, CNT-based soft actuator devices using heat [18–22], light [23–27], electric fields [28–31], magnetic fields [32–34], or an ion bias [35–37] have been reported; however, they have not yet been commonly used. One of the reasons is that their movement is simpler than that of the existing actuators. For this, we expected our CNT composite paper to be a candidate soft actuator and have developed our "paper actuator [38]". Since it would be made of paper, it is expected that complicated three-dimensional structures could be formed relatively easily and that movement with a high degree of freedom would be possible; however, some problems, e.g., operating performance, remain in the previous study. Therefore, we aim to improve the performance of our paper actuator in this study.

2. Materials and Methods

2.1. Structure and Operating Principle of a Paper Actuator

Figure 1 shows the structure of our soft actuator based on our CNT composite paper, i.e., the paper actuator. The actuator is an ion-conductive soft actuator and generates a bending motion in response to an electric field (input voltage). The actuator has a three-layer structure: two electrode layers and one electrolyte layer. The CNT composite paper is used as the electrode layers and ordinary paper containing ionic liquid is used as the electrolyte layer. It is known that the ionic liquid consists only of cations and anions.

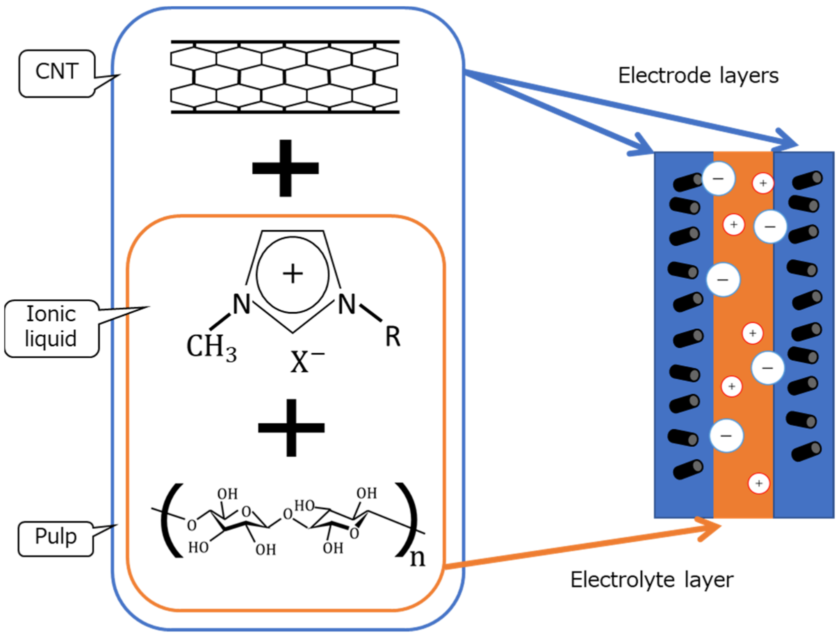


Figure 1. Structure of our paper actuator. Pulp is raw paper material (from Ref. [38] under a CC BY 4.0. license).

Figure 2 shows the operating principle of our paper actuator. When voltage is applied to the electrode layers, ions move and are injected into each electrode layer. At this moment, one side of the electrode layer stretches and the other side shrinks because the sizes of the cations and anions, which are components of the ionic liquid, are different. As a result, a bending motion is generated when one side stretches and the other side shrinks.

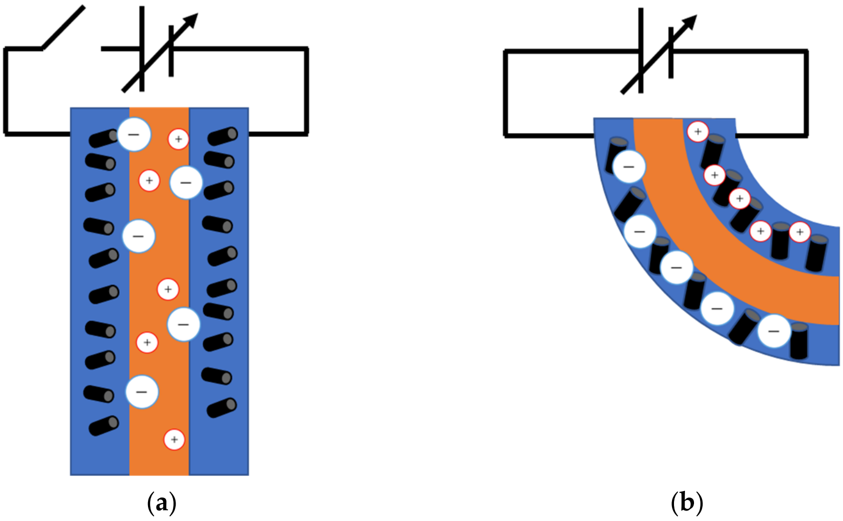


Figure 2. Operation of the paper actuator where the anion size is larger than that of the cations. (**a**) Before applying voltage. (**b**) After applying voltage (from Ref. [38] under a CC BY 4.0. license).

2.2. CNT Composite Paper Construction Method

CNT composite paper can be constructed in the following way based on the traditional Japanese *washi* method, as described in our previous study [13]. Figure 3 shows the construction process of CNT composite paper.

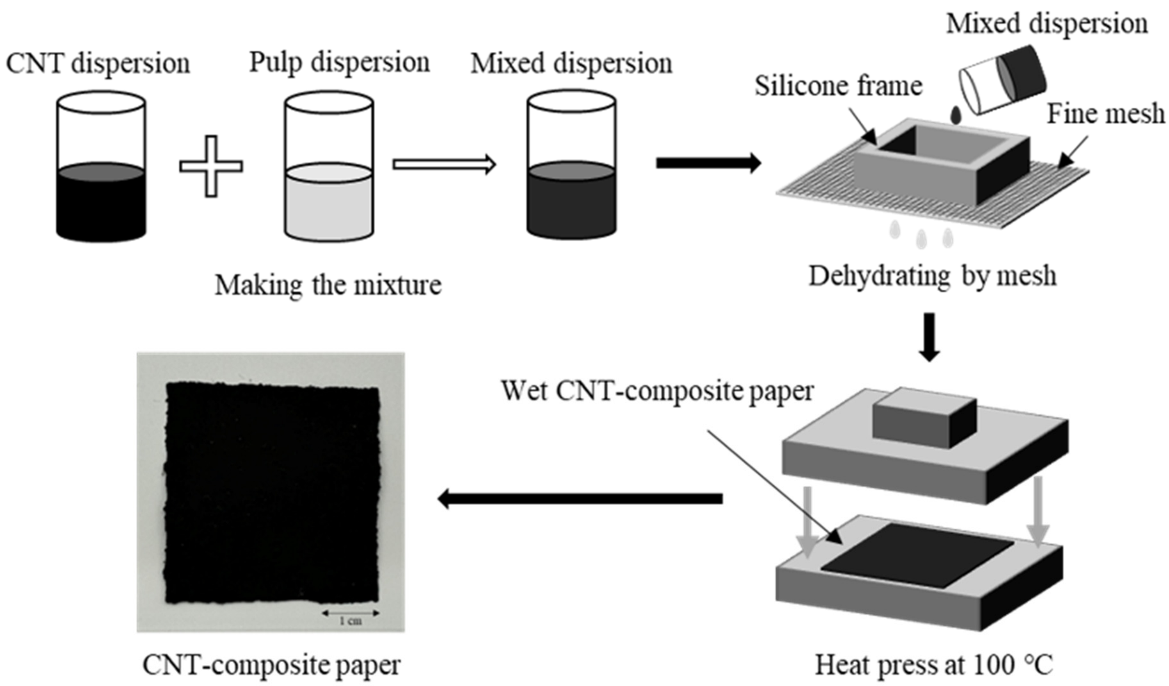


Figure 3. Schematic of the construction method for CNT composite paper.

As an example, the construction process of CNT composite paper is described as follows:

(1) Dispersing 10 mg of single-walled CNTs (ZEONANO SG101, Zeon Corporation, Tokyo, Japan) and 100 mg of catechin ((+)-Catechin Hydrate, NACALAI TESQUE, INC., Kyoto, Japan) as a dispersant in 10 mL of pure water with an ultrasonic homogenizer (UX-50, Mitsui Electric Co., Ltd., Tokyo, Japan) for 30 min.
(2) Dispersing 100 mg of pulp (raw paper material) in pure water with an agitator (SM-102, AS ONE CORPORATION, Osaka, Japan) for 30 min.

(3) Mixing the dispersions obtained in steps (1) and (2).
(4) Dehydrating the mixed dispersion by pouring it into a fine mesh for 1 min.
(5) Drying the wet CNT composite paper with a heat press machine (MNP-001, AS ONE CORPORATION, Osaka, Japan) at 100 °C for 1 h.

CNT composite paper can be fabricated by following the above process from step (1) to (5). Figure 4 shows the fabricated CNT composite paper. The mechanism of our paper actuator is similar to that of electrochemical energy storage; therefore, we have chosen SG101-CNT, which has a high specific area for our paper actuator.

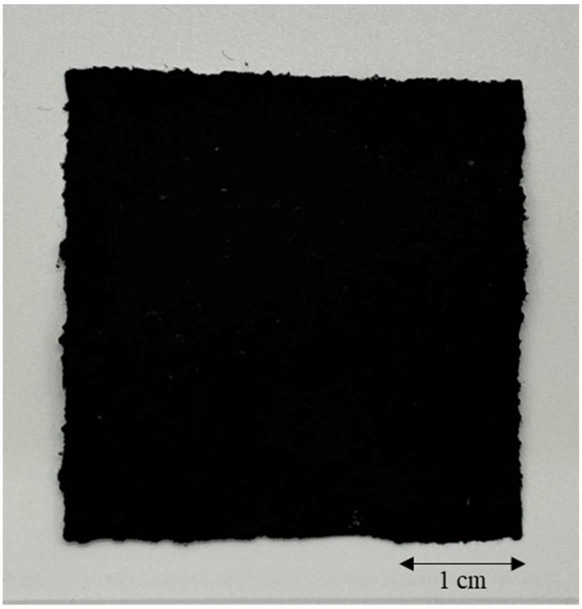


Figure 4. CNT composite paper.

2.3. Paper Actuator Construction Method

As described in Section 2.1, our paper actuator consists of three layers: two CNT composite papers as the electrode layers and one ordinary paper as the electrolyte layer. Figure 5 shows the construction process of the actuator.

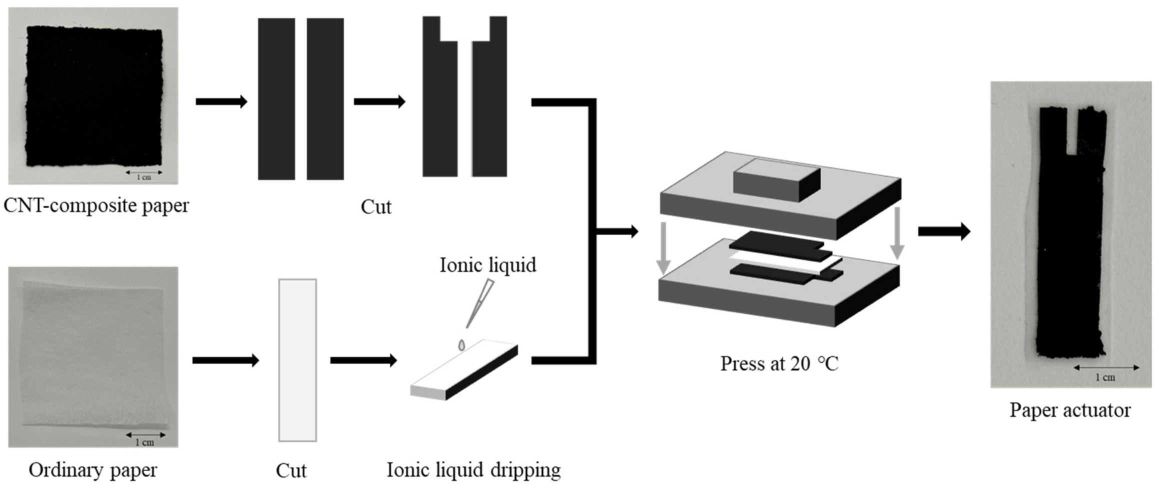


Figure 5. Schematic of the construction method for our paper actuator.

As an example, the construction process of the paper actuator using CNT composite paper is described as follows:

(1) Cutting the CNT composite paper into two 10 mm × 40 mm pieces with scissors.
(2) Cutting off a small piece from the edge of the CNT composite paper with scissors to make a jutted area for voltage input.
(3) Cutting ordinary paper (KAYDRY, NIPPON PAPER CRECIA CO., Ltd., Tokyo, Japan, in this study) with scissors into one 13 mm × 43 mm piece.
(4) Impregnating the piece of ordinary paper with 100 µL of ionic liquid (EMI-TFSI, Toyo Gosei Co., Ltd., Tokyo, Japan) for 3 h.
(5) Sandwiching one processed ordinary paper obtained in step (4) between two processed CNT composite papers obtained in step (2).
(6) Pressing them with the heat press machine at 20 °C and having them be stuck together for 10 s.

Our paper actuator can be constructed by following the above process from step (1) to (6). As described below, we added step (2) to modify our paper actuator in this study. Figure 6 shows the constructed actuator.

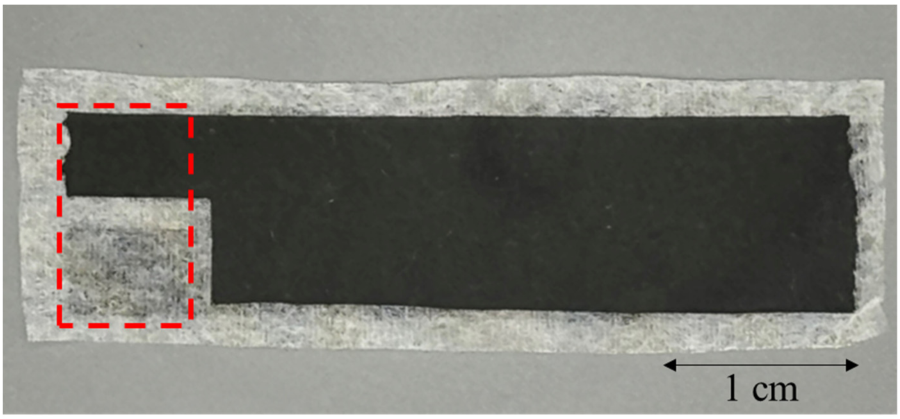


Figure 6. Constructed paper actuator. The dotted box indicates the area where the wooden clip for the voltage input clips in. For ease of viewing the structure, the actuator in this figure does not contain ionic liquid.

In our paper actuator, in order to prevent the electrodes from facing each other, the edge of the CNT composite papers used as the electrode layers, on the side where the voltage is input, is cut. This is to prevent the ions from moving at the edge of the CNT composite papers (around the input area). In this study, for simplicity, clips are used at the ends for voltage application and sample fixation. On the other hand, the clipping force may cause the ions to be pushed out of the paper. In addition, theoretically, the actuator would move due to the electric field generated on the facing surfaces, so the above device was used to prevent actuator motion from occurring at the edges.

2.4. Performance Evaluation Method of the Paper Actuator

Figure 7 shows the experimental setup for measuring the bending motion of our paper actuator. A wooden clip covering a graphene sheet is mounted on the edge of the actuator to apply the voltage and prevent the ionic liquid from leaking out of the actuator. When a rectangular wave voltage is applied to the actuator with an arbitrary function generator (FGX-2220, TEXIO TECHNOLOGY CORPORATION, Yokohama, Japan), the bending motion is measured with a laser displacement sensor (CD22-15V, OPTEX FA CO., LTD., Kyoto, Japan).

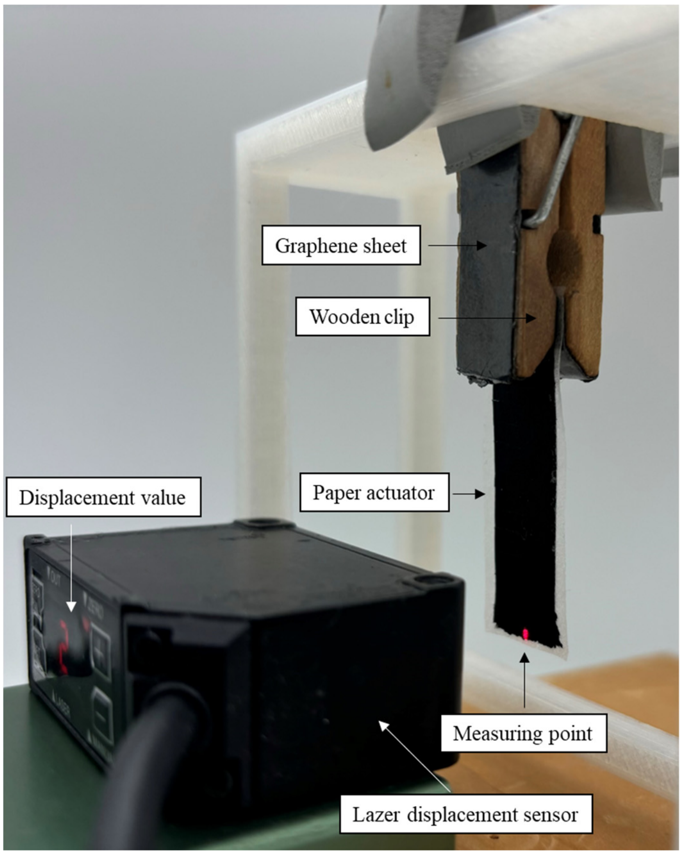


Figure 7. Experimental setup to measure bending motion.

3. Results and Discussion

3.1. Improvement of the Softness of CNT Composite Paper by Changing the Fabrication Conditions

In our previous studies, the bending motion of our paper actuator was so small that it was difficult to see. The reason for this was considered to be the stiffness of the actuator. In this study, we first tried to soften the actuator in order to improve the bending motion. For this, we focused on the CNT composite papers that were the electrodes of the actuator.

Generally, to soften paper, it is necessary to reduce the amount of pulp in the paper; however, in the case of CNT composite paper, reducing the pulp would cause problems such as the paper not being uniform or a piece of paper being missing in the configuration of CNT composite paper, making it impossible to make perfect CNT composite paper. Therefore, we attempted to reduce the amount of CNTs in the composite paper. It is known that CNTs in the composite paper are thought to have a role in connecting fiber to fiber [13,17,38], and it was confirmed that reducing the amount of CNTs effectively made the composite paper softer than the previous one in our preparatory experiments. To confirm the change in the composite paper in response to the change in the amount of CNTs contained, four samples of CNT composite paper were made and the softness of them was measured. In measuring the softness, one end of CNT composite paper was fixed with a clip to check the degree of deflection. In addition, it was generally known that the softness of the paper was affected by the thickness. We therefore also measured the thickness of the CNT composite papers with a digital outside micrometer (MCD130-25, Niigata Seiki Co., Ltd., Niigata, Japan). Figures 8 and 9 show the degree of deflection and thickness of the four prepared samples of CNT composite paper. As shown in Figure 8, there was little change in (**a**) and (**b**), while there was a large deflection in (**c**) and (**d**). As shown in Figure 9, the thickness of the CNT composite papers was proportional to the amount of CNTs.

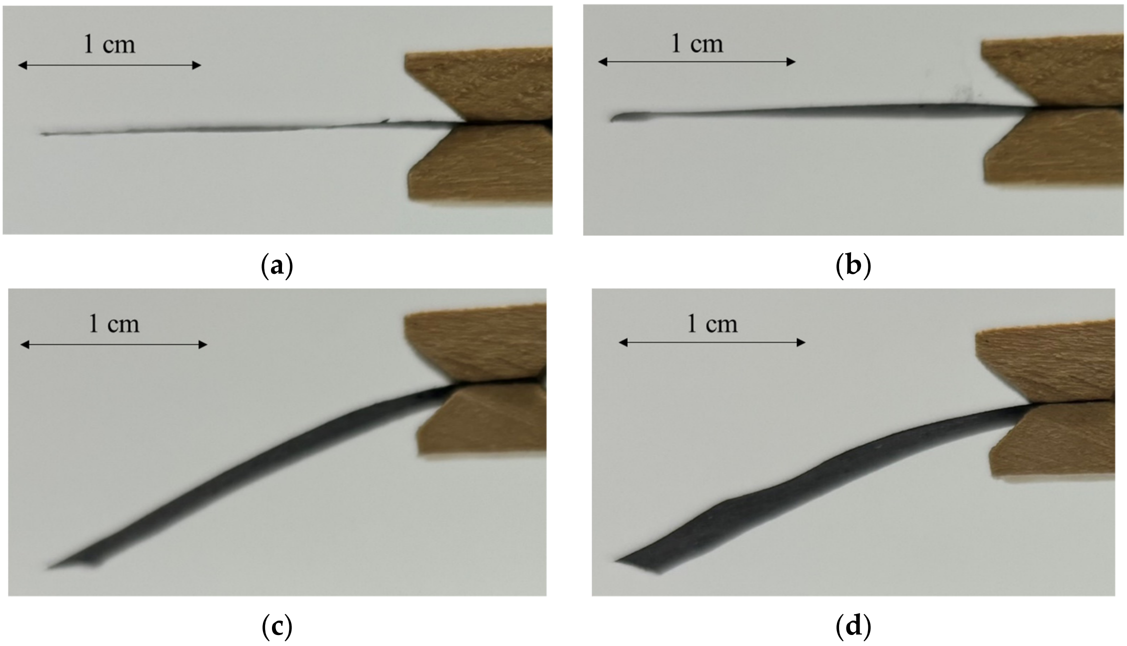


Figure 8. Difference in deflection depending on the amount of CNTs: (**a**) 30 mg, (**b**) 20 mg, (**c**) 10 mg, and (**d**) 5 mg.

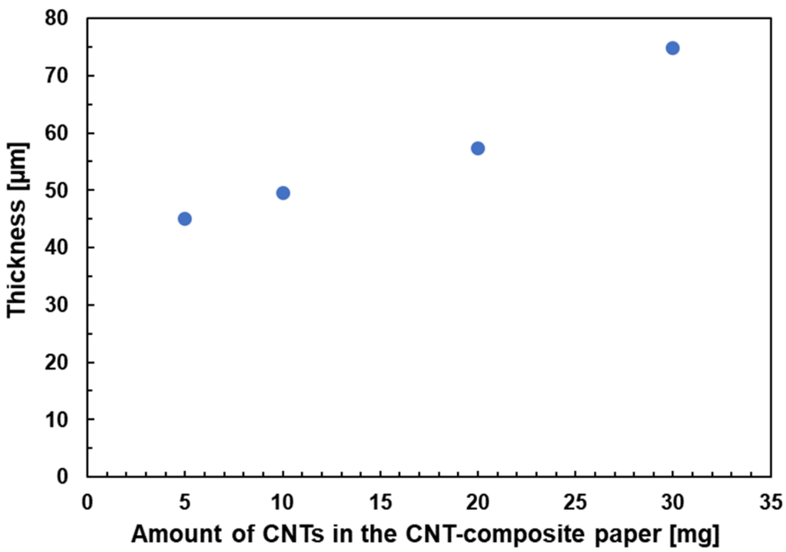


Figure 9. Difference in thickness depending on the amount of CNTs.

These results indicated that CNT composite paper became softer as the amount of CNTs was reduced. The slope of the thickness curve was gradual because the thickness was affected not only by the amount of CNTs but also by the force of the CNTs in connecting fibers to each other. In other words, the softness of CNT composite paper was affected not only by the amount of pulp but also by the amount of CNTs, and it was found that the amount of CNTs in the composite paper could be reduced from the previous conditions for CNT composite paper.

3.2. Actual Bending Motion of the Paper Actuator with Softened CNT Composite Paper

Based on the above improvements in our CNT composite paper, we constructed our paper actuator by following the construction process described above. Figure 10 shows the actual bending motion of the actuator when a rectangular wave voltage of ±3 V amplitude and 40 mHz frequency was input to the actuator. It was found that the bending motion of the actuator was confirmed to be sufficient. This bending motion was about 20 times greater than that of the previous actuator. As a result, it was found that softening CNT composite paper was very important for the bending motion of the actuator.

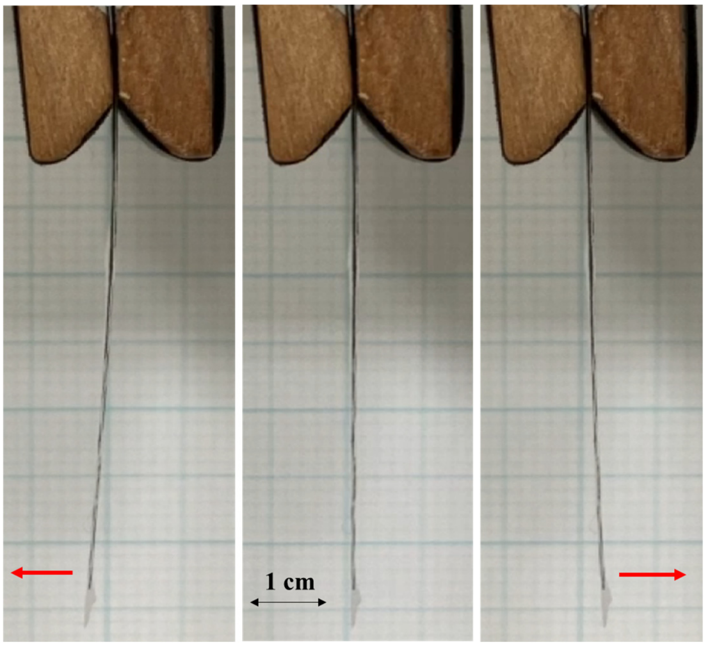


Figure 10. Actual bending motion of the paper actuator under a rectangular wave input voltage of ±3 V and frequency of 40 mHz.

3.3. Characterization of the Paper Actuator

Figure 11 shows the measured displacement, frequency response, response to voltage, and displacement duration when rectangular wave voltage was applied to our modified paper actuator. Figure 11a shows the displacement characteristic of the actuator under a rectangular wave input voltage of ±3 V and frequency of 40 mHz. The maximum displacement was 2 mm, which is 20 times greater than that of the previous actuator. Figure 11b shows the response of the actuator when the frequency of the input is changed under a rectangular wave input voltage of ±3 V. As a result, the displacement of the actuator followed the input frequency. In other words, when high-frequency rectangular wave voltage was applied to the actuator, the bending motion could not reach its maximum displacement; therefore, the displacement became smaller, resulting in a small oscillating motion. Figure 11c shows the motion of the actuator when the input frequency was fixed at 10 mHz and the input voltage was varied. It was found that the displacement of the actuator followed the input voltage; in other words, the ions in the actuator were moving under the influence of the input voltage. Figure 11d shows the motion duration characteristics of the actuator under a rectangular wave input voltage of ±3 V and a frequency of 100 mHz. It was found that the bending motion of the actuator continued for at least 1000 cycles. It has also been confirmed that the bending motion of the actuator continued for at least 2 days. The results show that the actuator works like other actuators; however, a gradual decrease in displacement was also observed. We consider that this was because the ionic liquid in the paper actuator moved downward via gravity, reducing the ions that affect the bending motion.

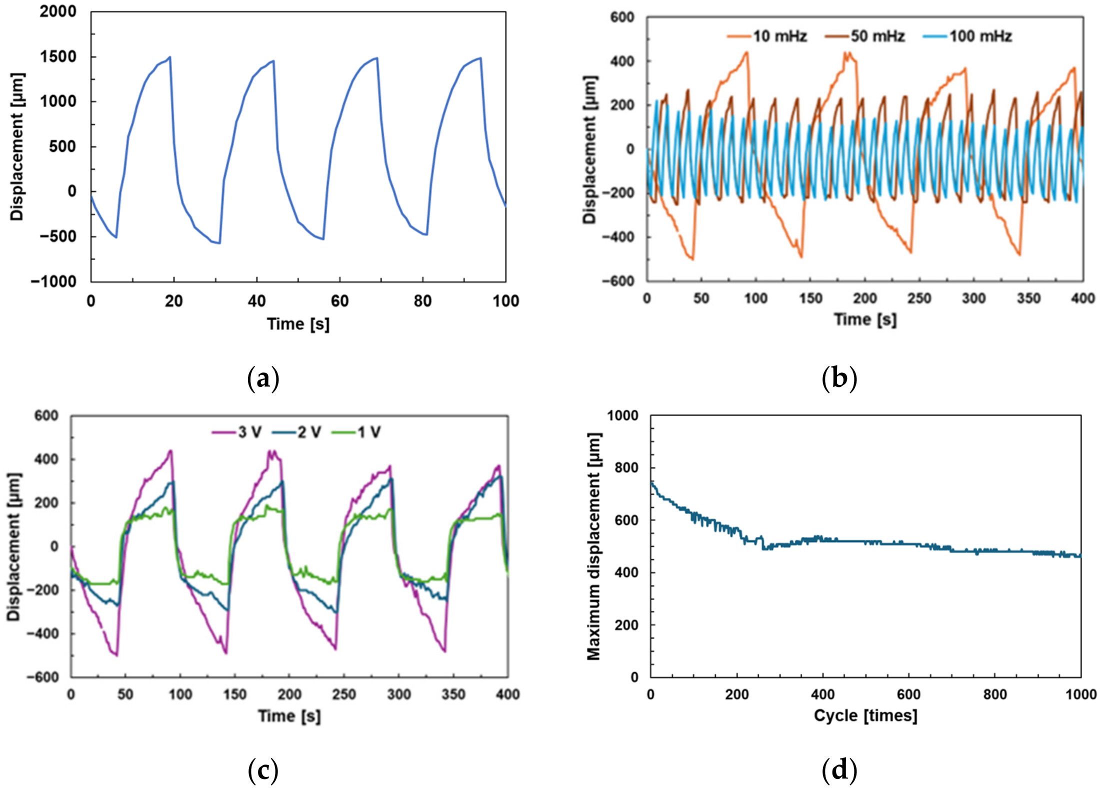
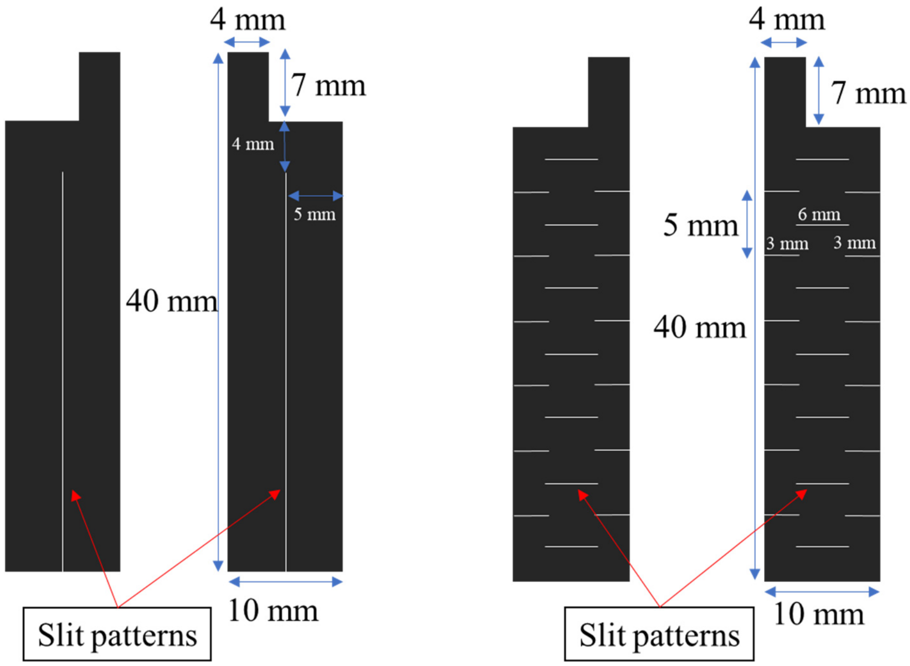


Figure 11. Chacterization of the paper actuator. (**a**) Displacement characteristics of the actuator under a rectangular wave input voltage of ±3 V and frequency of 40 mHz. (**b**) Displacement of the actuator with various frequencies under a rectangular wave input voltage of ±3 V. (**c**) Displacement of the actuator with various voltages under a rectangular wave at a frequency of 10 mHz. (**d**) Maximum displacement of the actuator under a rectangular wave input voltage of ±3 V and frequency of 100 mHz over 1000 cycles.

3.4. Kirigami Technique Introduces Paper Actuator

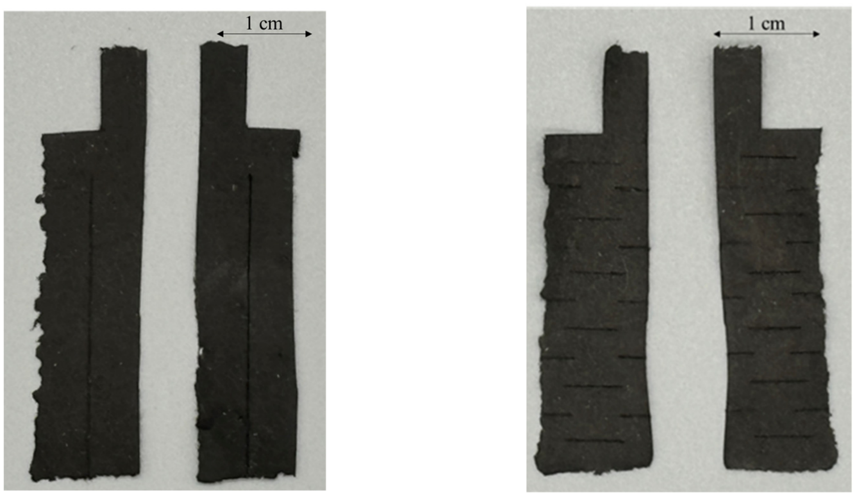
Finally, to obtain further softness, we examined the use of the *Kirigami* technique for our paper actuator, taking advantage of the fact that CNT composite paper is paper and its ease to process. The *Kirigami* technique is a Japanese traditional technique targeting papers, and a structure with periodical slits in the sheet is called a *Kirigami structure*. Paper with a *Kirigami* structure is known to show unique flexibility in the bending direction [39,40]; therefore, we believed that the introduction of the *Kirigami* technique could improve the bending motion of the actuator. In this study, for simplicity, we made periodic slits with a cutter knife in the CNT composite papers that were used as the electrodes of the actuator. Figure 12 shows the slit pattern of the CNT composite papers. The same slit pattern was incorporated in the other electrode.

Figure 13 shows the processed CNT composite papers and Figure 14 shows them clipped at the edge to determine the degree to which the sample deflects under gravity. It was found that CNT composite paper softens after the cutting processes. Using this sample, the response of the actuator when a voltage with an amplitude of ±3 V and a frequency of 10 mHz was given as the input is shown in Figure 15. The sample with a vertical incision shown in Figure 15a shows that the maximum displacement was 4 mm, which is greater than that without a vertical incision; therefore, it was confirmed that a vertical incision was effective for the bending motion. On the other hand, in the sample with a horizontal incision shown in Figure 15b, the maximum displacement was 1 mm, which did not differ much from the case without horizontal incision; however, the actuator was then found to perform not only a bending motion but also a twisting motion, as shown in Figure 16. This is thought to be due to the fact that the horizontal incision induced the electrode layer to deform from a two-dimensional direction to a three-dimensional direction, changing the bending motion into a twisting motion. Although the details were still under investigation, we presumed that the twisting motion was affected by the specific movement of the anions and cations in the ionic liquid, which was triggered by the specific slit patterns. We believe that the slits probably exhibit eye-pattern-like shape-changing behavior because of ion movement, which is the cause of the three-dimensional motion. Indeed, paper and sheet-like materials with such slits are known to easily form three-dimensional structures [41]. As a result, the twisting motion in addition to the bending motion was found to be feasible by incorporating the *Kirigami* technique into the CNT composite papers.



(**a**) (**b**)

Figure 12. Slit patterns of the CNT composite papers. (**a**) Vertical incision. (**b**) Horizontal incision.



(**a**) (**b**)

Figure 13. Actual processed CNT composite papers. (**a**) Vertical incision. (**b**) Horizontal incision.

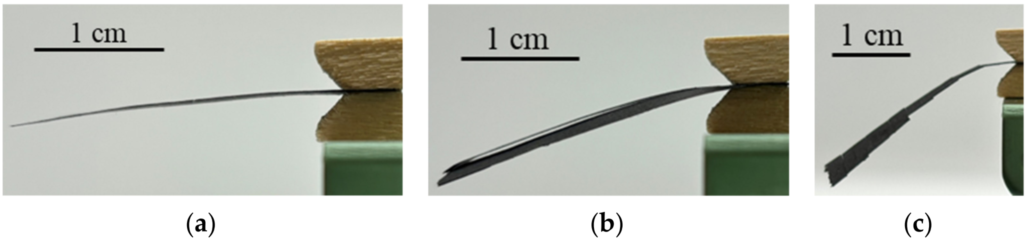


Figure 14. Deflection depending on the slit pattern. (**a**) No incision. (**b**) Vertical incision. (**c**) Horizontal incision.

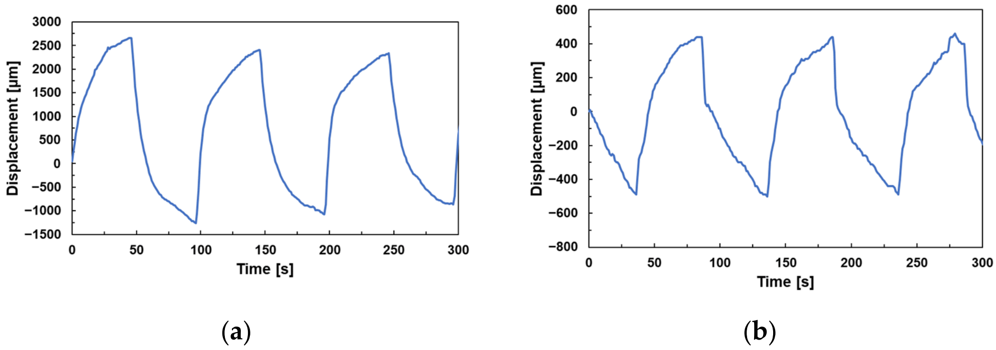


Figure 15. Displacement of the actuator under a rectangular wave input voltage of ±3 V and frequency of 10 mHz. (**a**) Vertical incision. (**b**) Horizontal incision.

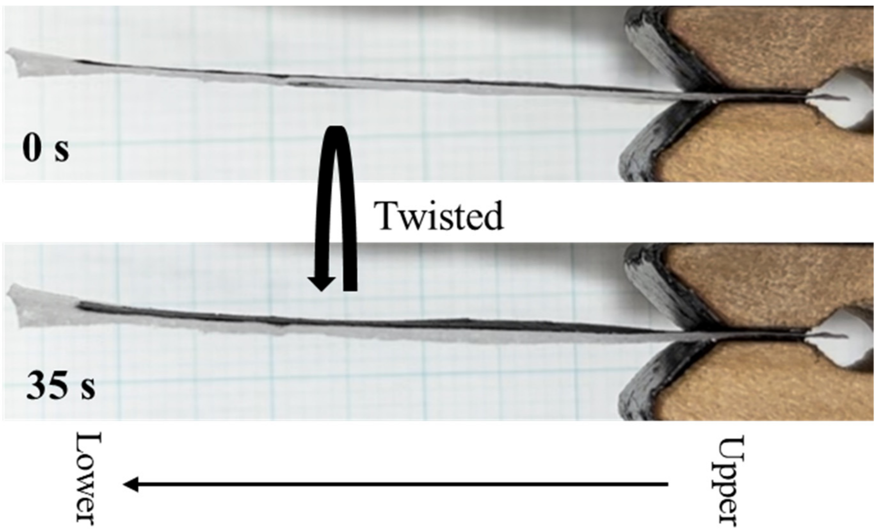


Figure 16. Twisting motion of the paper actuator with a horizontal incision. For ease of viewing, these figures are rotated 90° clockwise.

4. Conclusions

In this study, we aimed to improve the bending motion of our soft actuator, i.e., a paper actuator, using CNT composite papers. In our previous study, we demonstrated that actuator operation was possible by using CNT composite papers as electrodes with ordinary paper containing ionic liquid between the electrodes; however, its bending motion was not sufficient. Therefore, in this study, we attempted to improve the CNT composite paper, which is the electrode layer of the actuator, by softening it. To do this, we tried to reduce the ratio of CNTs contained in the composite paper. As a result, the CNT composite paper became softer, the performance of the actuator using it was improved, and its bending motion became visible.

The response of the improved actuator to the input voltage was investigated in detail, and the detailed operating conditions could be clarified for the first time. On the other hand, a problem was found regarding repetitive operation, in that the displacement became smaller with each repetition.

Then, as a new strategy, we took advantage of the fact that the proposed actuator was made of paper and introduced the *Kirigami* technique into the structure. As a result, it was found that not only a bending motion but also a twisting motion could be realized in specific slit patterns. It is thought that the fact that the variation in movement can be increased by simply adding incisions is unique to the proposed paper actuator. With the progress of future research, there are high expectations that this paper actuator will be put into practical use.

Author Contributions: Conceptualization, R.T. and T.O.; methodology, R.T.; validation, R.T. and T.O.; formal analysis, R.T.; investigation, R.T. and T.O.; resources, R.T. and T.O.; data curation, R.T.; model, R.T.; writing—original draft preparation, R.T.; writing—review and editing, T.O.; visualization, R.T. and T.O.; supervision, T.O.; funding acquisition, T.O.; project administration, T.O. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Conflicts of Interest: The authors declare no conflicts of interest.

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