All-solid-state carbon-nanotube-fiber-based finger-muscle and robotic gripper

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ABSTRACT
Carbon nanotube fibers (CNTFs) have many desirable properties such as lightweight, high strength, high conductivity, and long lifetimes. Coiled CNTF is an ideal material for preparing electrochemically driven artificial muscles. While previous studies focused mainly on the actuation performance of artificial muscles made of CNTF, this study focuses on an actuator that mimics human finger movements (flexion). More specifically, the preparation of CNTF muscles were optimized by twisting with weight. Then, actuators are designed and assembled by combining all-solid-state CNTF muscles with polypropylene (PP) sheets. Moreover, a dual-electrode system, which is infiltrated by a gel electrolyte, is built into the muscle actuator. In addition, a robotic gripper is fabricated, which uses these actuators. This study can help improve the design of CNTF-based muscle-actuators and future applications in robotics.
1. Introduction

In the human muscular system, muscle tissue can be categorized into one of three types: skeletal, cardiac, and smooth. Muscles typically work in pairs consisting of both flexors and extensors. The flexors contract and pull on the bone, which produces a movement of the joint. When the movement is completed, the flexors relax, and the extensors contract to either extend or straighten the bones. In other words, they can move limbs only via contraction and followed relaxation, they can never push bones back to their original position. For this reason, human palm muscles consist of antagonistically arranged flexors and extensors that enable the fingers to bend toward the palm. Today, artificial muscles are already available, which can be used to replace human muscles. It is also feasible to design flexible artificial muscle-structures that mimic the musculoskeletal system.

Inspired by natural muscles, various soft artificial muscles, made of natural or man-made fibers, were developed. Man-made fibers include synthetic polymer fibers, such as nylon fiber [1–6] and spandex fiber [7–11] as well as artificial inorganic fibers such as shape memory alloy fiber (SMAF) [12–18], graphene fiber [19–24] and carbon nanotube fiber (CNTF) [25–31]. Nylon fiber, spandex fiber, and SMAF are mostly actuated by heating with a heat gun or hot water. The actuation temperature of nylon fibers is about 250°C, and they feature good abrasion resistance, low price, and they can be produced using mature manufacturing technology. In addition, nylon fiber can be used on a large scale. The actuation temperature of spandex fiber is about 130°C. They are both acid- and alkali-resistant, corrosion resistant, and have a large actuation volume. Spandex fiber is usually used in smart wearable devices. SMAFs have a shape memory effect, which can generate a strong contraction force when the shape is restored. Unfortunately, their life cycle is short. SMAFs are commonly used in medical and aerospace applications. In addition, graphene-oxide fibers are very sensitive to wetness due to their large specific surface area and large number of functional groups. This means that their application environment is restricted. Compared with the above-mentioned fibers, CNTFs can be controlled through electrochemical reactions and they are lightweight, high-strength, with high electric conductivity, good thermal conductivity and flexible. These properties make CNTFs the preferred material for intelligent artificial muscles. Consequently, many types of CNTF twisted and coiled actuators (CTCAs), with contraction and torsion actuation, were reported.

There are still significant challenges for electrochemically actuated artificial muscles [32]. Recent studies have demonstrated that if a liquid electrolyte is used for ion-conduction, the weight and volume of the electrolyte and container exceeds that of CTCA by far. Researchers also worked on replacing artificial muscles, which were dipped in liquid electrolyte, with more practical all-solid muscles. Double-electrodes (with both winding and inner-outer braiding structures) are normally used for solid electrolytes [33]. The mechanically coupled working and counter electrodes are both actuating electrodes, so the mismatch between the stroke of the anode and cathode muscles can reduce muscle performance.

It was found that electrochemically-driven CNT muscles have a limited driving capacity [32,33]. Jae Ah Lee et al. fabricated all-solid-state artificial muscle yarns that could produce useful, stretched, muscle strokes (contraction of 10% at −10 V, 0.25 Hz, 26 MPa). Later, a muscle actuator with a concentric braided structure was able to support 37 MPa stress and contract 5% at −5 V by twisting 60 μm CNTFs in combination. Wang et al. [34]
produced quasi-solid-state yarn muscles, which consisted of PAN nanofiber-coated (isolated positive and negative electrodes) CNT-yarn working electrodes and bare CNT counter electrodes. The group used −4.5 V at 0.1 Hz and the muscles contracted by 18.34% under a stress of 8.5 MPa. Hence, it is necessary to find a good balance between driving capacity and twisting weight to ensure a sufficiently high electrocontractile capacity.

Inspired by the antagonistic arrangement structure of biological muscle systems, a new artificial muscle was developed in this study, where the CTCA acts as a flexor muscle and an elastic polypropylene (PP) sheet serves as extensor. A robotic gripper was also designed by assembling the artificial muscles. The framework for all studies is as follows: In Section 2, the preparation method and working conditions of CTCAs are introduced, and the characteristics of CTCAs are analyzed. In Section 3, the design of a PP sheet to improve the driving efficiency of the artificial muscle is explained. In addition, an artificial muscle actuation module was fabricated by combining the PP sheet and CTCA. In Section 4, the performance of the actuation module was tested. In Section 5, a robotic gripper was made by assembling the actuation modules. Finally, conclusions and future works were discussed in Section 6.

2. Preparation and characteristics of the CTCA

The performance of CTCAs depends greatly on their coiling structure. Although there are many preparation methods, it is difficult to produce CTCAs with excellent properties. In this study, the CTCA coiling quality was improved by attaching a weight at the end (during the twisting process). As shown in Figure 1), a CTCA is prepared by twisting a yarn made of 4 CNTFs (7 cm in length and 85 μm in diameter). The CNTFs used in this research were prepared by Hebei Tanyuan Nanotechnology Co., Ltd. One end of the CNTF yarn is fixed on the rotating rod, and the other end is loaded with a weight. Therefore, the axial force on the CNTF yarn helps avoid kinks during twisting. Several coils appear in the CNTF yarn during twisting, and the number increases until the yarn displays a ‘coil spring’ shape. Furthermore, multiple winding areas might occur simultaneously due to defects in the CNTFs – see Figure 1. Finally, coils are evenly distributed throughout the yarn and stacked on top of each other – see Figure 1). Therefore, the CTCA behaves like a coil spring. Subsequently, to enable the CTCA to contract (in response to electrical stimulation), the coils need to be pulled apart and held in this position. Therefore, the CTCA was stretched by attaching a heavier weight at the end. As shown in Figure 1), the CTCA, which was twisted with a 5 g weight, was stretched by 32% by a 10 g weight. After the CTCA had been stretched, the spring structure generated a restoring force that prevents it from deformation, and the restoring force increased with heavier weights.

The effect of the suspended weight on both twist-shortening and the extension of CTCAs were analysed. Figure 1) illustrates the shortening of CTCAs with different weights during twisting. In the absence of coils, the length decreased linearly with increased twisting, and in the presence of coils, the length decreased faster until the whole yarn was coiled. The heavier the suspension weight, the more twisting was required to achieve the same amount of shortening of the CTCAs. In the end, the length of each CTCA was reduced by more than 70%. The average coil diameter of a CTCA decreased with increasing weight – see Table 1. Overall, the coils were
evenly distributed across the CTCA, even though the defect resulted in few irregular winding structures. The extension for the CTCA with heavier weights is shown in Figure 1). The storage capacity of coiled CTCA yarn was increased with increasing weights and it was more difficult to stretch. In the following study, a 3 g weight was used for twisting, while a 5 g weight (about 13 MPa) was used to stretch the CTCAs. This ensured a suitable restoring force as well as adequate coil spacing for the CTCA.

When a single polarity voltage was applied to the CTCA in the electrolyte environment, the CTCA expanded in the radial direction and generated an untwisting torque in the twisted structure. The coils of the CTCA converted the untwisting torque into a contraction force along the length direction, which caused the CTCA to contract. The restoring force and the space between coils decreased with the contraction of the CTCA. The actuation started when the contraction force was equal to the sum of the restoring force and the weight. When the applied voltage dropped to zero, the CTCA returned to its original length and the restoring force (caused by the weight) was restored.

Figure 1. Preparation of the CTCAs: (a) Twisting of CNTFs, (b) CTCA coil spring, (c) Coils of CTCA are pulled apart by a weight; (d, e) Twisting and coiling characteristics for the CTCAs.
Table 1. Diameters of coiled CTCAs.

| Average diameter of coils | I  | II | III | IV |
|---------------------------|----|----|-----|----|
|                           | 0.517 mm | 0.481 mm | 0.432 mm | 0.393 mm |

3. Design and preparation of a flexion-extension actuator module

3.1. Design of the CMAM

A schematic of a palmar skeletal muscle and artificial flexion-extension structure is shown in Figure 2). In nature, muscles are fiber tissues that enable the bones to move using their traction. In this way, human fingers can perform a wide range of movements and coordinate to perform delicate movements, such as writing or playing the piano. The basic movements of fingers are flexion (motion toward the palm) and extension (motion away from the palm). The muscles that control the basic movements of each finger are called flexors and extensors. They consist of pairs of antagonistic muscles. Each finger is equipped with multiple flexors and extensors, which makes movements more delicate, and it becomes easier to distinguish strengths. When a finger needs to be flexed, the flexors contract, while the extensors are relatively relaxed. More specifically, the flexors contract and pull the phalanges, thus enabling finger flexion. Similarly, when a finger needs to be straightened, the extensors stretch, while the flexors are in a relatively relaxed state. Therefore, the finger muscles consist of an active actuator that can move in two directions. The flexors and extensors contract and pull the phalanges respectively, without interference. When the flexors and extensors are both in a relaxed state, the fingers are bent slightly in a ‘natural state.’ This mechanism not only controls the flexion and extension of fingers but also improves the efficiency of finger operation.

The flexor and extensor, which produce active movement only in one direction, are arranged with respect to each other such that a whole finger joint can rotate in both directions.

\[ F_{\text{muscle}} = F_{\text{flexor}} - F_{\text{extensor}} \]  

(1)

Here, \( F_{\text{muscle}} \) represents the net force of movement in the musculoskeletal system of fingers, \( F_{\text{flexor}} \) and \( F_{\text{extensor}} \) indicate the forces on the flexor and extensor, respectively.

If the extensor is in the relaxed state, \( F_{\text{extensor}} = 0 \), the force generated by the flexor is equal to the net force, i.e. \( F_{\text{muscle}} = F_{\text{flexor}} \), and the direction of the net force is the direction of movement of the flexor toward the palm. Usually, both flexors and extensors are contracted actively, and both the magnitude and direction of net force are determined by the force difference between flexors and extensors. When \( F_{\text{muscle}} = 0 \), it can be assumed that both flexors and extensors are in a relaxed state, i.e. \( F_{\text{flexor}} = F_{\text{extensor}} \). If \( F_{\text{flexor}} = F_{\text{extensor}} \neq 0 \), the finger can be considered to remain in a constant position with a certain degree of flexion or extension. Because flexors and extensors pull each other, the overall change of the finger position varies depending on the strength of the two muscles.
The CTCA-PP antagonistic pair were located at opposite sides, so two-directional active actuation became possible. This is illustrated by the simple force diagram shown in Figure 2).

\[ F_{\text{CMAM}} = F_C + F_{CE} - F_P \] (2)
Here, $F_{CMAM}$ represents the net force generated by the CTCA, $F_C$ represents the initial restoring force in the CTCA, $F_{CE}$ is the contraction force generated by the applied voltage, and $F_P$ represents the recovery force stored in the PP sheet when the actuator was extended.

In the antagonistic structure of the skeletal muscles of human fingers, when both the flexors and extensors were in a relaxed state, $F_{flexor} = F_{extensor} = 0$. This results in a skeletal muscle system that passively generates motion in response to external forces and cannot be restored even after the external forces had been withdrawn. However, this state of complete relaxation rarely occurs in practice, and this study focuses mainly on the non-relaxed state. In the CTCA-PP antagonistic pair, even the CTCA was not actuated, and any deformation of the structure (due to external forces) could be spontaneously restored after removing the external forces. This is consistent with $F_C = F_P \neq 0$. When $F_C + F_{CE} = F_P$, the CTCA is maintained for a certain degree of bending. $F_C$ is the initial load, which remained constant, which means $F_P$ increases with increasing $F_{CE}$.

By mimicking the skeletal muscles of human fingers, an electrochemically driven all-solid-state CNTF muscle-actuation module (CMAM) was developed in this study – see Figure 2. The CMAM consists of three parts: a PP sheet as extensor, a CTCA as flexor, and a printed circuit board (PCB) with a Pt sheet soldered as counter electrode. The PCB electrode was placed in the middle of the PP sheet with one short side taped on it. The other short side of PCB was free, which guides the directional deformation of the PP sheet. Two CTCAs (parallel with the PCB electrode) were glued to the PP sheet as working electrode. Between the CTCA and the PCB electrode, gel electrolyte was added to form a two-electrode structure. The two electrodes could move relative to each other without interference due to synchronous operation. In this way, the CTCA and the PP sheet act as antagonistic structures, enabling active activation in both directions [35–37]. Because the CTCAs were contracted by the voltage, the CMAM was bent by an angle.

As the CTCAs relaxed after the voltage turned zero, the PP sheet provided the recovery force to straighten the CMAM. However, the recovery force, which was generated by the PP sheet needed to be controlled within a certain range. It should not be too small to open the coils of the CTCAs, and not be too large to counteract the driving force of the CTCAs too much. Therefore, the dimensions of the PP sheet needed to be designed suitably. The thickness of the PP sheet was about 0.19 mm. We cut a 30 mm by 7 mm rectangular hole in the middle of the PP sheet. This length corresponded to the length of the CTCA coil part. The width of the side strips is denoted by $d$. The load-displacement characterization of the PP sheet was performed using an in-situ tensiometer, as shown in Figure 2). When $d$ is equal to 7 mm or 6 mm, the load increased with increasing displacement first and then decreased slowly after reaching a peak. When $d$ is equal to 4 mm or 5 mm, the load grew monotonically with increasing displacement until the highest value was reached. However, the PP sheet with $d$ equal to 4 mm was relatively soft and easy to deform when the CTCAs were stretched. After taking all the above into consideration, a PP sheet with $d$ equal to 5 mm was used to ensure sufficient reliability and efficiency of the CMAM.
3.2. Electric actuation mechanism

As shown in Figure 3, acetone and PVDF-co-HFP were mixed in an oil bath at 60°C for 2 hours to obtain 10 wt.% PVDF-co-HFP/acetone solution. TEA-BF4 was dissolved in PC to obtain 1 M electrolyte. The solutions of TEA-BF4/PC and PVDF-co-HFP/acetone were mixed in a beaker consistent with the required ratio (in this paper, the ratio of gel to electrolyte is 1:1), so that the two components mixed completely. The mixture was placed in a culture dish and left at room temperature for about 20 minutes to evaporate the acetone and gel the electrolyte [32,33,38].

The CTCA and PCB electrodes were spaced approximately 2 mm apart and filled with a solid electrolyte. This not only provided the ions needed for the system to form a circuit but also prevented the two electrodes from contacting directly and forming a short circuit. In this way, a two-electrode system (with a solid ionic gel as the electrolyte environment) as well as CTCA and PCB sheet as working electrode and counter electrode, respectively, were obtained. Conducting wires were attached to the CTCA, and the Pt electrodes, respectively, to input a square-wave voltage. The negative voltage at the CTCA electrode was compensated by the absorption of negative electrolyte ions within the circuit, while the positive voltage at the PCB electrode was compensated by the absorption of positive electrolyte ions within the circuit – see Figure 3. The CTCA muscle served essentially as a supercapacitor (as described in the literature), and its contraction was actuated by the migration of ions from the surrounding electrolyte into the electrochemical bilayer of the CNTFs [6,33]. When voltage was applied to the clockwise twisted CNTFs, the CTCA begin to untwist due to electrochemically induced radial expansion – see Figure 3. Furthermore, the coil structure of the CTCA converted the untwisting torque into a contraction force [33,35,39]. During the contraction of the CTCA, there were also some coils untwisted to compensate for the diameter of the remaining coils.

4. Actuation performance of the CMAM

The actuation performance of the CMAM was studied. The experimental device is shown in Figure 4. The weight of the actuation module was roughly 2 g. A hole was made in the middle of each end of the PP sheet. We attached one end of the PP sheet to the iron stand and attached a weight via the steel ring at the other end (with the PCB electrode placed vertically). Conductive wires connected the CTCA and PCB electrodes to a waveform generator (Keysight 33500B series waveform generator, designed and manufactured by Keysight). Different weights were suspended on the actuation module, and different voltages were applied. Both contraction and deflection of the CMAM were studied. All measurements were performed three times.

4.1. Contraction performance

The contraction performance of the CTCA is shown in Figure 4. Weights of 5 g, 10 g, and 20 g were attached to the CMAM, respectively. A square-wave voltage (with the amplitudes −4 V, −6 V, −8 V and −10 V) was applied between the two electrodes, respectively. When the CTCA were actuated for a longer period, the contraction accumulated more. In order to maximize the deformation of the CMAM, the frequency was set to 0.01 Hz. The
contraction of the CTCAs tended to linear for 5 g weights. When the voltage amplitude was −10 V, the CTCAs, which were suspended with a weight of 5 g, contracted by 15.2%. When the suspension weights reached 10 g and 20 g, the contraction performance of the CTCAs decreased substantially at low voltage. However, when the voltage reached 10 V, the shrinkage reached 13.3% and 9.6%, respectively. The CTCAs could lift a weight about 2500 times of their own weight (0.0118 g). This confirms that these CTCAs showed better contraction characteristics than previously reported muscle yarns [33] that were actuated in solid electrolyte environments.

4.2. Deflection performance of the CMAM

The deflection performance of the actuators is shown in Figure 4). The PP sheet was bent by the contraction of CTCA yarns. By applying voltages, the PP sheet could be deflected by angles ranging from 29° to 67°. As the hanging weight and the voltage increased, the PP sheet bent by increasing angles. The deflection of the CMAM was analyzed for two cycles, when the suspended weight was 10 g. The applied stress (26
Figure 4. (a) The actuating characteristics of CMAM; (b) Contraction performance of a CTCA muscle; (c) Deflection angle for different weights; (d) Angle-time curve for the square-wave voltage and one cycle; (e) The actuation cycles of CMAM.
MPa) was 63 times higher than the capability of natural skeletal muscle [32]. As shown in Figure 4), the frequency was 0.01 Hz, and the duty cycle was 50%. During the former half cycle, a smooth angular deflection occurred at both −6 V and −8 V. When the voltage was −10 V, the deflection rate tended to increase and then decrease, until the maximum deflection angle was reached. In the second half of the cycle, the angular deflection dropped rapidly and then slowed to approach the initial value. However, because both the contraction and relaxation of the muscle yarn relied on the changes of the helical structure, sometimes these changes were irreversible. Therefore, the deflection angle of the PP sheet could not be fully returned to the original value after actuation. Figure 4) shows 300 actuation cycles of the CMAM with a 20 g load. An average deflection angle of 1.9° is resulted during these cycles, which were actuated by a square-wave voltage pulse with frequency of 0.1 Hz and voltage of −10 V (50% duty cycle). The creep was below 1%.

5. A robotic gripper based on CMAMs

A robotic gripper was developed by combining three CMAMs with a holder – see Figure 5. The actuator is attached to the root of the clamp. First, the clamp held the end of actuator, which made it possible to adjust the angle of the actuator via the gap. Then, the hot-melt adhesive was filled into the gap, i.e. the actuator was attached completely without movement. Furthermore, the hanger of the holder was threaded into the tail of the clamp. After adjusting the distance from the clamp to the center of the holder, the tail of the clamp was attached using the hot-melt adhesive. The dimensions of the robotic gripper are shown in Figure 5) and its specification parameters are shown in Table 2. A platform was built for the robotic gripper to grab the object – see Figure 5). The device consisted of an iron stand, a pulley, a soft plastic strip, weights, ball (radius of 20 mm, weight of 8 g), and an electronic balance. The robotic gripper was attached to a soft plastic strip, which was connected to a weight at the other end via a pulley. In this way, the robotic gripper could be moved up and down by moving the weight. Conductive wires connected the positive and negative terminals of the mechanical gripper to the function signal generator. Care was taken to avoid short circuits.

5.1. Gripping performance of the robotic gripper

First, the contraction of the CTCA flexor muscles was driven by a square-wave with a fifty percent duty cycle, −8 V, and 0.0125 Hz, which enabled the 3 CMAMs to bend. When L was smaller than the radius of the ball, the 3 free ends were close together and could grip the ball – see Figure 6). Then, the robotic gripper was slowly lifted by a weight, while the 3 CMAM fingers remained in proximity. Next, to exhibit the extension of the extensor muscle. As shown in Figure 6, the weight was moved left and the lifted robotic gripper was dropped slowly.

The process used in the experiment should match the deflection characteristics of the actuator- see Figure 6, and the three grippers were still approaching when the robotic gripper was down-see supporting information (movie 2). The robotic gripper from the
beginning of the drive, grip, up and down, the time control within 40s. In this way, when the robotic gripper landed, the robotic gripper extension muscle could be extended spontaneously and smoothly put down the weight—see Figure 6).

6. Conclusion

Flexor and extensor muscles, which (in humans) contract in response to nerve impulses, form antagonistic structures in the musculoskeletal system of human fingers. This enables the fingers to move. Similarly, CTCA, which is made of coiled CNTFs, can contract in response to an applied voltage. In this study, a novel actuation module was prepared that used an antagonistic structure consisting of CTCA and PP sheet. When a balance between CTCA and PP sheet was reached, the PP sheet gently pulled the coil structure of the CTCA

![Figure 5. Dimensions (a), and mass (b) of the robotic gripper; (c) Robotic gripper setup.](image)

![Table 2. Specifications of the robotic gripper.](table)
To study practical applications, a robotic gripper was developed by combining three CMAMs with a frame. The manipulating gripper was very light, with a total mass of only 18.5 g. Thanks to the cooperation of the three-actuator module, the manipulator could successfully perform a gripping action. Appropriately increasing the number of CNTFs in CTCAs could increase the carrying capacity of the robotic gripper and make it more versatile. In future studies, it would be desirable to optimize the solid electrolyte to make it easier to coat. This could reduce the amount of electrolyte needed and decrease
voltage loss. In addition, the heat from the Pt sheet accelerated the consumption of solid electrolyte, which could be improved in the future. In addition, for future artificial muscle actuators, encapsulation will also be important. Just like a muscle needs skin for protection, the design of a suitable flexible shell could be another topic for improvement.

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Disclosure statement

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