Origami hand for soft robotics driven by thermally controlled polymeric fiber actuators

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Abstract

Active fibers can serve as artificial muscles in robotics or components of smart textiles. Here, we present an origami hand robot, where single fibers control the reversible movement of the fingers. A recovery/contracting force of 0.2 N with a work capacity of 0.175 kJ kg⁻¹ was observed in crosslinked poly[ethylene-co-(vinyl acetate)] (cPEVA) fibers, which could enable the bending movement of the fingers by contraction upon heating. The reversible opening of the fingers was attributed to a combination of elastic recovery force of the origami structure and crystallization-induced elongation of the fibers upon cooling.

Introduction

Hands have a very delicate and complex structure compacting tendons, muscles, nerve fibers, blood vessels, and bones in precise positions.^[1, 2] This allows muscles and joints a great range of movement to fulfill various tasks spanning from grasping objects to lifting things or from catching a ball to threading a fine needle. The motion of the fingers of hands is controlled by tendons, which are connected to agonist–antagonist muscle pairs located in lower arm responsible for the actual movement by their contraction and expansion. These tendons slide in the tendon sheath to transmit the muscle movement, where the fluid in the sheath ensures smooth sliding of the tendons. In the past few decades, researches have been inspired by natural hand in order to design functional grippers for robotic applications.^[3, 4] Different approaches have been followed to recreate and study multifingered robotic hands while developing dexterous manipulation in order to build robotic systems, which can not only interact more safely and conveniently with humans and environment but also perform some tasks.^[5–7] Conventional technologies such as electromagnetic, pneumatic, hydraulic, or fluidic have been explored as actuating systems for robotics.^[8–10] Rigid-bodied actuators such as electric motors or low-profile actuators such as shape-memory alloy (SMA) wires have been applied and studied extensively to provide natural hand-like movements in these robotic systems.^[11, 12] However, despite the advantages of SMA over electric motors, such as high force to weight ratio, compact size, or noiseless operation, drawbacks including limited strain, hysteresis, and high cost restrict their deployment as actuators on a larger scale.^[4, 13]

Flexibility and compliance offered by soft polymeric material-based actuators allow to implement pre-designed motions,

offer easy deformability and adaptation to changes in the environment.^[14, 15] These actuators are rapidly emerging at the frontier of technological innovations due to their wide array of applications in humanoid robots, artificial muscles, energy harvesting materials, or production technologies (e.g., as micro-grippers).^[16–18] Special attention has been paid to develop fiber actuators, which allow complex actuation behaviors based on their flexibility and high anisotropic properties.^[19] Soft polymeric fibers can also act as an actuator, where a specific thermomechanical treatment including deformation by twisting, stretching, or bending could implement the actuation capability to the fibers.^[19, 20] Recent breakthrough in this regard was the discovery of tensile and torsional fiber actuators based on low-cost fishing line and sewing thread.^[21, 22] These fiber actuators were further exploited to achieve thermally or electrothermally driven muscles and were deployed in a 3D-printed hand model.^[4] The advantages of these actuators are the larger stroke/actuation with minimal or no hysteresis compared with large hysteresis and small actuation of SMAs. Similar strategy has been used to develop an electro-thermally driven robotic hand from metal-coated sewing threads as artificial muscles.^[23] In these approaches, the fiber muscles were deployed as agonist–antagonist pair similar to the natural system, where the movement of fiber acting as an agonist can be reversed by the contraction of the second fiber, which acts as antagonist.

Crosslinked networks based on semi-crystalline polymers are capable of free-standing reversible actuation and are suitable for application where a reversible muscle-like movement should be realized by a single actuator.^[24, 25] Radiation crosslinked fibers have been recently reported for their thermally triggered torsional and electrically triggered tensile actuation.^[26, 27] The

Karl Kratz—deceased.

free-standing reversible actuation relied on melt-induced contraction (MIC) of the oriented crystallites on heating and crystalline-induced elongation (CIE) on cooling. In this study, we demonstrate an origami robotic hand, in which the movement of the fingers can be triggered by crosslinked poly[ethylene-*co*-(vinyl acetate)] (PEVA)-based fibers actuating in heating channels, see Fig. 1. We hypothesized that reversible movement of the fingers can be realized by deploying single-fiber actuators in contrast to the approaches where fibers as an agonist–antagonist pair are required. The fibers contract and expand reversibly in response to heat, whereas this motion is transmitted to (move) the fingers via guiding fibers, which act as tendons in our system. The contraction of the fiber on heating led to bending of the finger. The challenge was to achieve opening of the fingers without deploying an additional (counter) fiber actuator. Our strategy to achieve the reversible finger movement was a combined response of elongation of the fiber on cooling and elasticity of the origami structure. The elasticity of the origami hand fingers was investigated and compared with the recovery force of the fiber actuators and related work capacity. The effect of crosslink density on the recovery force and work capacity of the fibers was further explored (by varying irradiation dose for crosslinking). Furthermore, the effect of irradiation dose on the thermomechanical as well as actuation properties of these fibers was investigated in this study.

Materials and methods

Materials

Poly[ethylene-*co*-(vinyl acetate)] (PEVA) with a vinyl acetate content of 18 and 28 wt% (Elvax® 460A and 260A, DuPont, Delaware, USA), triallyl isocyanurate (TAIC) (99%, Sigma-Aldrich, Steinheim, Germany), and toluene (99.8% Sigma-Aldrich, Steinheim, Germany) were used without any purification.

Methods

Fiber preparation and crosslinking

Fibers were prepared from the blend granulates (PEVA + TAIC) as described in the references.^[26, 27] These fibers were then irradiated by gamma beam for different doses, such as 99, 132, and 165 kGy. Sample IDs were given based on the VA content of the polymer and irradiation dose, for example, cPEVA18-099, cPEVA18-132, and cPEVA18-165 for cPEVA18 fibers, while cPEVA28-099, cPEVA28-132, and cPEVA28-165 for cPEVA28 fibers.

Fabrication of the actuating origami hand

The origami hand model was cut from printed paper and the fingers were folded to replicate natural hand's metacarpophalangeal (MCP) joints. A small piece of polypropylene tube 7×5 mm (length and diameter) was mounted on each joint section to guide the fibers. Finally, the tips of the fingers were connected to the fibers, which were directed through the guiding tubes into the heating channels. For the ease of the design, continuous longer fibers were used which act as actuating fiber (the part in the heating channels) and the guiding fiber (the part outside the heating channels) simultaneously. The heating channels were custom made 30.0×1.5 cm (length and diameter) from polypropylene test tubes (see supporting Fig. S1). The fibers were heated to $\sim 65^\circ\text{C}$ by blowing hot air into the heating channels by heat guns, while the temperature was monitored by a sensor. Cooling to room temperature $\sim 25^\circ\text{C}$ was achieved by simply switching OFF the heat guns.

Swelling experiments

The crosslinked fibers were analyzed by swelling experiments in toluene at 85°C where the extraction time was 24 h followed by 24 h for solvent evaporation in a vacuum oven at 45°C . The

Figure 1. (a) Schematic illustration of natural hand showing tendons (gray lines), tendon sheath (blue), bones (yellow), and muscles (orange) in the forearm. The closing and opening of the fingers are triggered by the expansion and contraction of the muscles, while tendon transmits these motions. Here, the muscles are always present agonist–antagonist pairs to achieve reversible movements. (b) Schematic illustration of origami hand model, where each finger is connected to a single fiber, which is responsible for the reversible actuation of the fingers by thermally triggered actuation in the heating channels. The red and blue color represents the heating and cooling of the channels, respectively.

gelation degree (G) was calculated from the isolated weight (m_{iso}) of the fibers and the dry weight (m_d) after extraction using Eq. 1.^[28] Non-extracted fibers were employed for thermomechanical and actuation investigations of the fibers.

$$G = \frac{m_d}{m_{\text{iso}}} * 100\%. \quad (1)$$

Differential scanning calorimetry (DSC)

DSC measurements were performed on a calorimeter (Netzsch, Selb, Germany) DSC 204, with heating and cooling rates of 10 K·min⁻¹. Thermal data reported correspond to the second heating, whereas the area under the melting and crystallization peaks led to calculate the corresponding enthalpies. Crystallinity related to the sample weight is calculated using Eq. 2.

$$\chi_c = \frac{\Delta H_m}{\Delta H^\circ m/100} * 100, \quad (2)$$

where $\Delta H^\circ m/100$ is enthalpy of melting of 100% crystalline polymer, which is 287 J·g⁻¹ for polyethylene.^[29]

Determination of elastic properties

Mechanical properties of crosslinked fibers at ambient temperature were assessed by tensile tests on a Zwick Z2.5 (Zwick, Ulm, Germany) with a strain rate of 5 mm min⁻¹. Young's modulus (E) and elongation at break (ϵ_{break}) were obtained and analyzed using 0.4 × 30.0 mm (diameter, length) fiber samples.

Quantification of reversible actuation

Cyclic actuation experiments were performed on Zwick Z1.0 (Zwick, Ulm, Germany) equipped with a thermochamber and temperature controller (Eurotherm Regler, Limburg, Germany). These tests consisted of five consecutive heating and cooling cycles (without programming module) under stress-free conditions. The fibers were reversibly heated and cooled between $T_{\text{low}} = 10^\circ\text{C}$ and $T_{\text{high}} = 65$ (for cPEVA28 fibers) or 75°C (for cPEVA18 fibers) with 3 K·min⁻¹, and the change in strain was recorded. An equilibration time of 10 min was used in each cycle at T_{low} and T_{high} . Quantification includes reversible actuation/strain (ϵ'_{rev}) and is calculated from Eq. 3, where $l_{T,\text{high}}$ and $l_{T,\text{low}}$ are the lengths at T_{high} and T_{low} , respectively, in reversibility cycles.

$$\epsilon'_{\text{rev}} = \frac{l_{T,\text{low}} - l_{T,\text{high}}}{l_{T,\text{high}}} * 100. \quad (3)$$

Contracting force of the fibers

Recovery or contracting force in cPEVA fibers was investigated from strain-controlled cyclic heating-cooling experiments using the same setup. The fibers were held in clamps with fixed strain, and change in force was recorded with change in temperature. These experiments consisted of five cycles, whereas in each cycle, the samples were heated to 65°C followed by an equilibration time of 10 min and cooling to 25°C with subsequent waiting time of another 10 min. The heating and cooling

rates were 3 K min⁻¹. The work capacity per weight of the fibers can be calculated by dividing the work by the mass of the actuating fiber as equation.^[30]

$$\text{Work capacity} = \frac{F \cdot \Delta L}{m_f}, \quad (4)$$

where F is the maximum contracting force of the fibers (at ~65°C) in N, ΔL is the change in length in meters during reversible actuation, and m_f is the weight of the fiber in kg.

Elastic force of the fingers

The force required to pull the fingers was measured on a Zwick Z1.0 tensile tester (Zwick, Ulm, Germany) where the fingers were connected individually to the strings and reversibly pulled and relaxed to a similar extent as observed in the origami hand demonstrator from thermally actuating fiber. Similarly, these experiments consisted of five cycles, whereas in each cycle, the strings were pulled to 20 mm elongation followed by release to 0 mm with a strain rate of 5 mm min⁻¹ and the change in force was recorded.

Results and discussion

The thermally actuated tensile actuation of cPEVA18-165 and cPEVA28-165 was investigated where the thermal properties had been already determined and are listed in reference.^[26] A higher $\epsilon'_{\text{rev}} = 10 \pm 1\%$ could be observed for cPEVA28-165, while the reversible actuation of cPEVA18-165 was only $\epsilon'_{\text{rev}} = 2 \pm 0.5\%$ without post-programming and under stress-free conditions. Furthermore, a cyclic test with 25 reversibility cycles was performed to confirm the suitability of these fibers (cPEVA28-165) as actuators. Here, a slight change of 1–2% in the reversible strain was found within the initial cycles, which stabilized after 10 cycles as shown in Fig. 2(a). The reversible actuation related to the extrusion-induced orientation of crystallizable segments in such fibers is demonstrated in an origami style hand robot [Fig. 2(b)]. The fingers of such an origami hand were connected with guiding fibers to one actuating cPEVA28-165 fiber, which was repeatedly heated and cooled in a custom made setup as shown in Video S1 and Fig. S1. Closing of the fingers by contraction of the fiber during heating is clearly visible. Fingers open reversibly upon cooling. The opening of the fingers is a result of elastic recovery of the origami structure and the elongation of the fiber actuators upon cooling, which allowed the backward movement.

In order to investigate the elastic force of the origami structure, which contributes to the opening of the fingers during reverse movement, cyclic tests were performed. Here, the origami fingers were attached to inextensible strings and reversibly pulled and released in a tensile tester to record the change in the force. The force was recorded over an extension from 0 to 20 mm, which led to the bending of the finger to a similar degree as it was observed during actuation movement of the fingers during thermal actuation. The maximum force was

Figure 2. (a) Cyclic test with 25 reversibility cycles showing the thermally controlled reversible actuation for cPEVA28-165. (b) Thermally controlled movement of selected fingers of demonstrator hand.

0.1 ± 0.01 N, which was observed at 20 mm extension and full finger folding as presented in Fig. 3(a) for the middle finger. This force was measured for all the fingers individually and results show a negligible change ~ 0.01 N in force for different fingers. Upon release, a fast drop of the force is observed initially to ~ 0.3 N with initial release to 17 mm followed by a slow decrease in force to 0 N with further release to 0 mm. The force at 20 mm represent the force required to hold the finger in the bend position, which drops down quickly upon initial release as the finger is allowed to relax. The force required to bend the thumb was comparatively higher 0.12 ± 0.01 N, which might be attributed to the higher friction as result of higher bending angle/degree of the pulling string.

Furthermore, the recovery force of the fiber actuators was investigated. The cyclic tests with constant strain were performed where the change in force during heating and cooling

was recorded. The cPEVA28-165 fibers were initially investigated, as they resulted to be more suitable for their reversible actuation performance and were deployed as actuators/muscles in our origami hand model. Identical temperatures (to the actuating hand) such as 25 and 65°C were selected here for these measurements. These tests are with constant strain so no change in length is observed. The measurement was started below crystallization temperature; therefore, the fiber is expected to be in a fully extended state, which on heating should try to contract. The results showed a force of ~ 0.2 N at 65°C [see Fig. 3(b)], which is the recovery/contraction force of these fibers related to entropic recovery of the polymeric chains upon heating. In the cooling cycle, the crystallization-induced elongation is expected; however, due to fixed strain, no further elongation can be observed. Instead, there is a gradual decrease in force, which is related to the CIE well as to the

Figure 3. (a) Elastic force of the fingers as a change in force during pulling and relaxing the middle finger. (b) Recovery/contracting force of cPEVA28-165 fiber.

decreased mobility of the polymer chain. When compared, this recovery/contraction force generated by the fibers is larger than the force required to pull the fingers. From this force, the work capacity $\sim 0.175 \text{ kJ kg}^{-1}$ was estimated for cPEVA28-165 fibers, which is the work capacity of fibers under stress-free condition (without any load applied).

The recovery force and the related work capacity can be influenced by the degree of crosslinking of the fibers; therefore, fibers with various crosslink densities were further explored in this work. The fibers were irradiated by gamma beam of different intensities such as 99, 132, and 165 kGy. Therefore, in addition to the change in recovery force, these fibers based on different PEVAs (PEVA18 and PEVA 28) as well as different irradiation doses were investigated for change in crosslink densities, thermomechanical properties, and for change in actuation performance. The swelling experiments revealed high gel content $G=95\pm 2\%$ for cPEVA28-165, whereas a decrease in $G=91\pm 1\%$ was found for lower irradiation dose fiber cPEVA28-099. A similar trend in G was observed for cPEVA18 fibers; here, G decreased from $90\pm 1\%$ to $86\pm 2\%$ (see Table I). A broad melting transition related to the crystalline PE domains was found for all cPEVA fibers. In case of cPEVA18 fibers, this melting transition was in the range between 50 and 90°C with a melting temperature interval of $\Delta T_m=40^\circ\text{C}$ and melting peak maxima around $T_m=81\pm 2^\circ\text{C}$, see Table I. The cPEVA28 fibers (with higher VA content) exhibited lower melting $T_m=66\pm 2^\circ\text{C}$ as well as crystallization $T_c=43\pm 2^\circ\text{C}$ temperatures; however, the melting transition was similarly broad but ranged from 40 to 80°C . Overall, the thermal properties were not much influenced by changes in the irradiation dose for the crosslinking.

Significant differences in the melting enthalpies (ΔH_m) were observed. Here, cPEVA18 fibers exhibited higher $\Delta H_m=75\pm 4 \text{ J g}^{-1}$ compared to cPEVA28 fibers $\Delta H_m=49\pm 2 \text{ J g}^{-1}$. Consequently, the resulting weight%

crystallinities (χ_c) calculated from ΔH_m were higher for cPEVA18 fibers $\chi_c=26\pm 2\%$. The lower $\chi_c=17\pm 1\%$ of cPEVA28 could be attributed to the lower crystalline polyethylene (PE) and higher amorphous vinyl acetate (VA) content in the starting material of these fibers. In general, the ΔH_m and χ_c of the fibers were considerably higher when compared with starting bulk materials, which might be attributed to the crystallite alignment in the extrusion process during the fiber synthesis. Besides the substantial impact of PE and VA content on the thermal properties, the variation of the irradiation dose results only in minor changes, where T_c , T_m , ΔH_m , and χ_c slightly decreased in with higher intensity irradiated fibers (see Table I), while ΔT_m remained unchanged. Tensile tests at ambient temperature revealed a significantly higher Young's modulus $E=80\pm 5 \text{ MPa}$ for cPEVA18-165 fibers with slightly reduced elongation at break $\epsilon_b=490\pm 30\%$. An increase in $\epsilon_b=570\pm 35\%$ with a decrease in $E=63\pm 3 \text{ MPa}$ was observed for the fibers with lowest irradiation dose cPEVA18-099. A similar trend in E and ϵ_b was found for cPEVA28 fibers; however, $E=47\pm 6 \text{ MPa}$ was small for these fibers as compared to cPEVA18 fibers, see Table I.

After the thermomechanical investigations, the parameters such as the cyclic temperatures can be defined. Since there was no significant change in thermal properties with change in irradiation dose, similar temperatures of 25 and 65°C were used in cyclic investigations of the recovery force for cPEVA28-099 and cPEVA28-132 using the same test protocol as for cPEVA28-165. The results revealed showed similar recovery force $\sim 0.2 \text{ N}$ for the fibers with 099 and 132 kGy. The identical recovery force might be attributed to the similar molecular chain alignment, which was solely achieved during extrusion process of the fibers, whereas the post crosslinking of the fibers had no effect on the recovery force of the fibers. Furthermore, cyclic thermomechanical tests were performed to explore the actuation capability of the fibers

Table I. Thermomechanical characteristics of cPEVA18 and cPEVA28 fibers.

Composition ^a	G^b [%]	T_m^c [$^\circ\text{C}$]	T_c^c [$^\circ\text{C}$]	ΔH_m^d [J/g]	χ_c^e [%]	E^f [MPa]	ϵ_b^g [%]	ϵ'_{rev}^h [%]
cPEVA18-099	91 ± 1	82 ± 1	62 ± 1	72	25	63 ± 3	570 ± 35	9 ± 1
cPEVA18-132	93 ± 1	82 ± 1	61 ± 1	75	26	79 ± 2	540 ± 25	10 ± 1
cPEVA18-165	95 ± 2	81 ± 1	60 ± 1	80	27.7	80 ± 5	490 ± 30	11 ± 1
cPEVA28-099	86 ± 2	67 ± 1	44 ± 1	48	16.7	42 ± 5	570 ± 15	2 ± 1
cPEVA28-132	87 ± 2	66 ± 1	43 ± 1	49	17	47 ± 2	430 ± 35	2 ± 1
cPEVA28-165	90 ± 1	65 ± 1	43 ± 1	50	17.5	53 ± 3	410 ± 20	3 ± 1

^aComposition: Sample IDs were given based on different VA contents of two PEVAs and different irradiation doses.

^b G : Gel content determined by swelling experiments.

^c T_m and T_c : Melting and crystallization temperature peaks determined from DSC measurements.

^d ΔH_m : Total melting enthalpy calculated as the area under the curve of the melting process.

^e χ_c : Weight percent crystallinity of PE calculated from DSC data.

^f E : Young's modulus determined by tensile tests at ambient temperature.

^g ϵ_b : Elongation at break during tensile tests at ambient temperature.

^h ϵ'_{rev} : Reversible strain determined from cyclic experiments.

crosslinked with various doses of gamma radiation. These tests consisted of only heating and cooling cycles under stress-free conditions without any programming step as described. Cyclic temperatures such as higher and lower temperatures in reversibility cycles (T_{high} and T_{low}) were selected based on the obtained thermal properties. A $T_{\text{high}} = 65^{\circ}\text{C}$ within the melting range was preferred for cPEVA28 fibers, while $T_{\text{low}} = 10^{\circ}\text{C}$ below the crystallization peak temperature was chosen. However, for cPEVA18 fibers, higher $T_{\text{low}} = 25^{\circ}\text{C}$ and $T_{\text{high}} = 75^{\circ}\text{C}$ were specified, taking into account their higher melting and crystallization ranges. The results of such cyclic tests are presented in Table 1. Here, the first cycle was considered as a training cycle, and the presented results are based on the average and standard deviation of reversible strain from second to fifth cycle. Reversible strains up to $10 \pm 2\%$ could be obtained in cPEVA28, while lower reversible strains $2 \pm 1\%$ were found for cPEVA18 fibers. However, it is interesting to observe that both fibers could show reversible bidirectional actuation without programming. Furthermore, the effect of irradiation dose was not significant on reversible actuation performance, and a negligible change in reversible actuation 1–2% was observed with decrease in irradiation dose in the fibers. Similar reversible strains of the fibers crosslinked with lower doses of gamma beam irradiations (99 and 132 kGy) might be the consequence of similar degree of orientation of the crystallites achieved during extrusion process.

Conclusion

In this study, we demonstrate an origami robotic hand where the movement can be initiated/controlled by thermally driven polymeric fibers in the heating channels. A single fiber was deployed as a reversible actuator, which on contraction upon heating led to the bending of the fingers. The opening of the fingers on the other hand was attributed to the combination of fiber elongation on cooling and hand elasticity. A recovery force of 0.2 N was observed for cPEVA28 fibers, which was double when compared to elastic force of the hand fingers. The work capacity of 0.175 kJ kg^{-1} was discovered for the fibers in free-standing condition. A maximum tensile actuation of $\sim 10\%$ was observed for cPEVA28 fibers, while a smaller actuation $\sim 2\%$ was perceived for cPEVA18 fibers. The reversible actuation was solely attributed to the crystallite orientation implemented in extrusion process. In addition, the effect of irradiation dose on the thermomechanical as well as actuation performance was investigated in detail. Here, an increase in crosslink density and a decrease in elongation at break is observed with increase in irradiation dose; however, the effect of crosslink density (or irradiation dose) was not that significant on reversible actuation performance of the fibers.

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Data availability

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Conflict of interest

M.B., K.K., and A.L. are co-inventors of patents and patent applications in the field of shape-memory polymer actuators.

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Supplementary Information

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