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# Artificial muscles based on coiled UHMWPE fibers with shape memory effect

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**Abstract.** Ultra-high molecular weight polyethylene (UHMWPE) fibers drawn at drawing ratio of 6 (pre-deformation strain 500%) demonstrating the obtained one-way shape memory effect. Artificial muscles have been manufactured in the form of coiled UHMWPE fibers. Isometric recovery stress and recovery strain of the fibers were measured during heating by using a dynamic mechanical analyzer (DMA). As a result, the fibers were capable to demonstrate large contraction of 78% (recovery strain of 93%) due to the entropic elasticity. The recovery stresses of the fibers reach up to 27 MPa. The work of stroke cycle for coiled artificial muscles with a constant stress of 1 MPa was recorded. Artificial muscles based on coiled UHMWPE fibers have a large stroke of 64 %. The structural mechanisms of muscle-like behavior were discussed.

***Keywords:*** *smart polymers, UHMWPE, artificial muscles, recovery stress, stroke*

## 1. Introduction

Development of artificial muscles is an important and a challenging goal for modern materials science. Artificial muscles have advantages in comparison with traditional drives, such as noiselessness, smooth running, compact dimensions, high capacity and a wide variety of activation methods (heat, electric current, electromagnetic field, chemical reaction, etc.). Application areas are extensive: robotics, exoskeletons, micro-drives, prostheses, smart sensors.

Ideal artificial muscles should have the following properties: high capacity, high work efficiency, large stroke, fast response, lightweight, hysteresis-free actuation and low cost. Various materials, such as carbon nanotubes [1–3], shape metal alloys (SMA) [4, 5], electroactive polymers [6, 7], and polymers with shape memory effect [8, 9] are used for manufacturing of artificial muscles. Unfortunately, the highly efficient artificial muscles are still successfully obtained but at a high cost.

The significant advantage of shape memory polymers (SMP) is the largest attainable strain. The deformation of a SMP can reach up to 100–1000% and more, while SMAs can be deformed only by 6–8% [10]. But SMP have a lower recovery stress compared to metals: 150 MPa for SMP [11] against 300–900 MPa for SMA [10]. However, the artificial muscles based on SMP will have much less weight because the density of SMP is 5–7 times less than the density of SMA. The specific recovery stress of SMPs is comparable, and sometimes exceeds the specific recovery stress of SMAs.

Recent achievements are represented by highly efficient artificial muscles made of nylon fibers and braided polyethylene [12], which have reversible contraction at temperature variations. Such artificial muscles being lightweight have a large stroke of more than 30% at high stress that finally allows generating more than 2000 J/kg of specific work at muscle contraction. These activators are distinguished

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by their manufacturing simplicity and have a low cost. The fabrication of artificial muscles made of nylon 6 was reported in [13, 14], which provide high thermo-mechanical properties and tensile stroke reaching to 50%. The mechanisms of artificial muscle stroke were discussed using the model of negative linear expansion coefficient in highly oriented amorphous-crystalline fiber precursors [15], or considering the geometric parameters of the artificial muscles, such as precursor fiber diameter, coil index (the ratio of the coil diameter to the precursor fiber diameter) and unloaded coil bias angle ($\alpha_c$) [12–15].

In this study, first we described the large stroke of coiled artificial muscles made of drawn UHMWPE (drawing ratio (*DR*) of 6). UHMWPE is a thermo-responsive SMP. Above the melting point, the polymer completely recovers its shape. The net-point in UHMWPE has a physical nature (entanglement of macromolecules). UHMWPE has a pronounced one-way shape memory effect (SME) with high recovery stresses [16] that we have utilized as the basic working principle for creating coiled artificial muscles.

The functioning of artificial muscles in mechanical devices presumes intimate surface contact with neighboring artificial muscle groups or other parts of the device. The contact during the movement of a muscle leads to wear and premature failure. UHMWPE has unique antifriction properties [17, 18] (low coefficient of friction, high wear resistance), which can facilitate the creation of wear-resistant artificial UHMWPE-based muscles. UHMWPE also has almost zero water absorption, which allows the use of UHMWPE-based artificial muscles to be operated in an aqueous media or under hydrothermal activation of artificial muscles. Therefore, we forecast a particular interest in the coiled UHMWPE artificial muscles by the engineering society.

## 2. Experimental details

UHMWPE provided by Boreskov Institute of Catalysis (Siberian Branch of Russian Academy of Science) having an average molecular weight of $4 \cdot 10^6$ g/mol and a high drawability was used to form the fibers. UHMWPE xerogel fibers were prepared by means of extrusion of UHMWPE/P-xylene gel through a 1 mm diameter die in a ram extruder at a temperature of 140 °C. The P-xylene solvent was used as a plasticizer of UHMWPE, which also leads to the formation of a microporous structure that helps to promote the orientation of the polymer. The concentration of P-xylene was 2.5 ml per 1 g of UHMWPE. The resulting xerogel fibers were dried for 48 h at 50 °C. The fabrication of oriented xerogel fibers was carried out through the uniaxial drawing at a temperature of 110 °C targeting a final *DR* of 6 (pre-deformation strain of 500%). In the experiment it has been found that the drawing temperature of 110 °C allows obtaining high orientation of UHMWPE fibers after drawing. As a result, UHMWPE fibers with a diameter of 400 μm were obtained.

In order to prepare coiled artificial muscles, UHMWPE fibers were twisted according to the technique described in [12, 13]. Bottom tip of UHMWPE fiber was fixed and loaded with a constant force. Another tip was twisted about its axis until getting coiled, Figure 1a. While twisting, UHMWPE fiber was loaded with a nominal stress of 10 MPa, which was defined as the ratio of the applied load over the cross section of the fiber before twisting. The overpassing of 10 MPa threshold resulted in the reduction of the coil

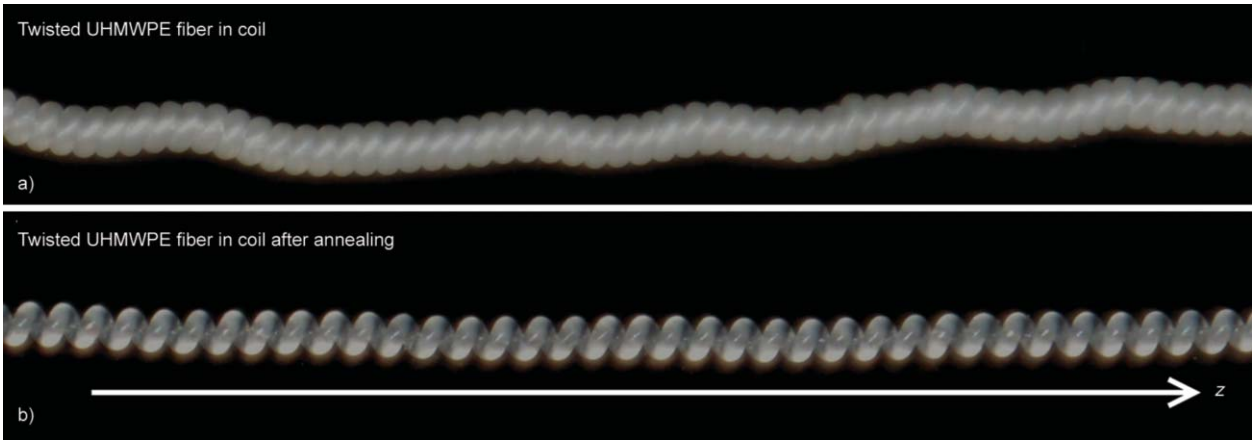


**Figure 1.** UHMWPE fibers after twisting (a) and further annealing (b).

index and, therefore, stiffer coiled fiber. Lower load made coiling almost impossible. Thus the applied stress of 10 MPa was chosen to form coiled fibers with the maximal coil index. According to [12] the increase in coil index leads to a larger stroke of artificial coil muscles. Development of artificial muscles with maximal actuation stroke was the main target in the present study.

The endpoint of the melting peak of UHMWPE fibers with *DR* 6 was at 147 ºC (see Figure 2), so this temperature was chosen for complete annealing of the twisted fibers. UHMWPE fibers were annealed by means of the immersion into the silicone oil at 147 °C for 30 sec. Both tips of the coiled fiber were fixed to prevent rotation, but still being free to move along the *Z* axis. After the cooling of the annealed coiled fibers down to the ambient temperature the self-stretching was observed. Annealed coiled fibers remained stretched without load at a room temperature, Figure 1b. Finally, artificial muscles made of coiled UHMWPE fibers were obtained with the diameter of 930 μm and coil index of 2.1.

Thermal analysis of xerogel fibers, fibers with *DR* 6 and artificial muscles were carried out by differential scanning calorimeter (DSC) NETZSCH DSC 204 F1 (NETZSCH, Selb, Germany) in argon atmosphere according to ASTM D 3417–83. The measurements were performed under controlled heating and cooling regime at the rate of 10 °C/min. The relative degree of crystallinity was calculated as the ratio of the experimental sample melting enthalpy compared to the completely crystallized polyethylene melting enthalpy, which was equal to 293 J/g [19]. The artificial muscle had a coiled shape. For ensuring good thermal contact between aluminum crucible and the material, the artificial muscle was cut into small pieces. The size of the pieces was around 0.5–1 mm. The pieces of artificial muscle were evenly distributed on the bottom of aluminum crucible to achieve a good thermal contact.

The fibers and coiled artificial muscles were investigated by using DMA Q800 (TA Instruments, USA) in tension mode at a frequency of 1 Hz, and were heated starting from room temperature up to 160 °C with a rate of 10 °C/min. Minimum initial force and oscillation amplitude for both types of samples were set 0.1 N and 10 μm, respectively. The length of samples between clamps was 15 mm.

Drawn UHMWPE fibers and coiled UHMWPE artificial muscles were studied using the DMA Q800 dynamic mechanical analyzer. Several test series were conducted in order to characterize thermo-mechanical properties presented in the following chapters.

### 2.1. Isometric recovery stress of UHMWPE fibers and coiled UHMWPE artificial muscles

The purpose of this experiment was to determine the maximum recovery stress. Both ends of the sample were fixed in clamps, so the distance between the clamps did not change during the experiment. Samples were heated starting from ambient temperature up to 180 °C at 10 °C/min. The heating process has activated the sample force returning to its permanent shape before the thermal orientation drawing.

### 2.2. Recovery strain of UHMWPE fibers during heating

Recovery strain characterizes the ability of the material to reversibly return to its initial shape under external stimulus. Drawn UHMWPE fibers were heated starting from ambient temperature up to 180 °C at 10 °C/min. One tip of the sample was fixed in a stationary clamp and the other one was fixed in a freely moving clamp with a minimal preload force of 0.001 N. The fiber recovery strain was calculated as shown in Equation (1):

$$\frac{L_0 - L_r}{L_0 - L_i} \cdot 100\% \tag{1}$$

where $L_0$ – length of a drawn fiber before heating, $L_r$ – length of a recovering fiber that is being changed upon heating, $L_i$ – initial length of a fiber (length of the fiber before drawing by thermal orientation).

### 2.3. Stroke cycle work of artificial muscle at a constant stress

One tip of the artificial muscle was fixed in a stationary clamp and the other one was fixed in a freely moving clamp at a constant stress of 1 MPa. Artificial muscles were heated starting from 50 °C up to 160 °C at 10 °C/min, and hold for 5 min, and then cooled down to 50 °C at 10 °C/min. Artificial muscle stroke was calculated according to Equation (2):

$$\text{stroke} = \frac{L_{\text{initial}} - L_{\text{final}}}{L_{\text{initial}}} \cdot 100\% \tag{2}$$

where $L_{\text{initial}}$ – length of artificial muscle before heating, $L_{\text{final}}$ – length of artificial muscle after heating.

## 3. Results and discussion

Negative linear expansion in highly oriented amorphous-crystalline precursor fibers is considered as the main driving force for the stroke of coiled artificial muscles [12–15]. For nylon fibers, the reversible contraction during heating can reach up to 4% due to the negative thermal expansion, and the same is for polyethylene fibers, which is about 0.3% [12]. The geometric parameters of the coiled artificial muscles, such as precursor fiber diameter, coil index and unloaded coil bias angle, $\alpha_c$, are also discussed as a governing factor of artificial muscle stroke [12].

On the other hand, polymer materials with shape memory effect (SME) can be deformed for hundreds of percent and further controllably contracted during heating due to the entropic elasticity [16]. We have intentionally studied the properties of the artificial muscles fabricated of polymer fibers manifesting the one-way SME, and we have expected that the properties of the artificial muscles depend on the fibers' recovery stresses and contraction during heating.

Figure 2 shows DSC curves for xerogel fibers, drawn fibers and UHMWPE-based artificial muscles. The xerogel fibers have crystallinity of 56% and the melting temperature in the range of 127–143 °C with the peak at 138.6 °C, Figure 2a. As a result of orientation drawing, the crystallinity of fibers increases up to 68%, and the range of melting shifts to higher temperatures, Figure 2b. The melting temperature of polymers mainly depends on the size of crystallites. Small and defected crystallites possess lower melting temperature as compared to larger size and defect-free crystallites. Such a melting behavior is related to the surface energy change in accordance with Thomson-Gibbs equation [17, 20]. The increase in crystallinity in the oriented drawn fibers is induced by the orientation of macromolecules and their recrystallization that is commonly observed phenomenon for highly oriented fiber and films [21, 22].

The annealing of coiled fibers in artificial muscles does not change the crystallinity, but leads to a shift in the onset and the maximum of melting peak to the higher temperatures, Figure 2c. The shift of the melting point is related to the reduction of the crystalline phase defects density.

Figure 1b shows that the annealing causes the stabilization of the coils shape and increases the distance between coils in artificial muscles. Moreover, the increased distance between coils in the artificial muscles enables the high stroke. We suppose that the annealing

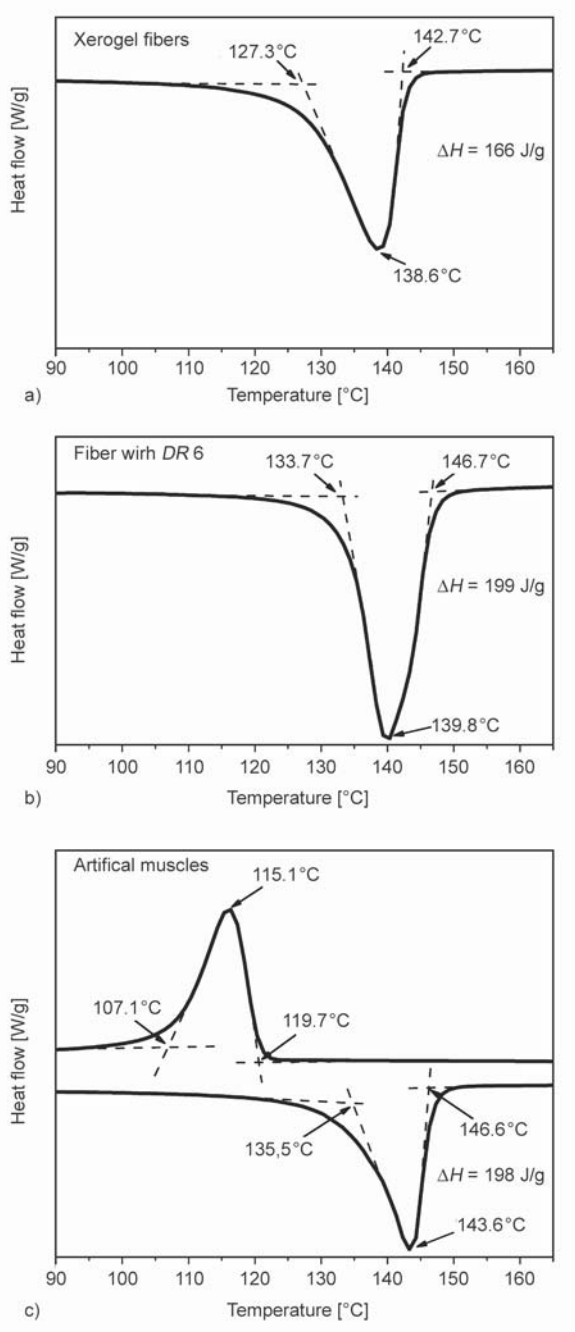


**Figure 2.** Typical DSC curves for a) xerogel fibers, b) fibers with *DR* 6 and c) artificial muscles.

releases residual internal compressive stresses inherited from the twisting and coiling deformation.

Figure 3 shows almost a gradual increase in the isometric recovery stress of drawn UHMWPE fibers upon heating. The maximum stress was 27 MPa at 160 °C. Further heating causes the rupture of the sample due to the exceeding of the recovery stresses over the strength at this temperature. The recovery stress obtained for the drawn UHMWPE fibers is rather high, and it exceeds the measured recovery stresses of the commercial highly oriented UHMWPE fibers

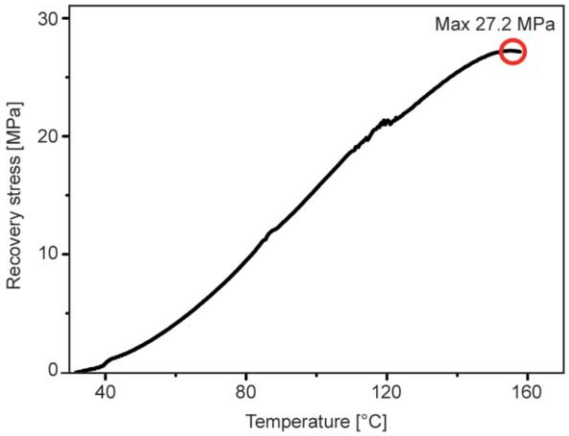


**Figure 3.** Isometric recovery stress of the drawn UHMWPE fibers during heating.

(recovery stress 22 MPa) obtained by gel forming method [16]. It has been argued that the value of the recovery stress depends on a degree of polymer chain elongation, because the polymer chains behave like a spring [16], so it can be expected that more oriented (with higher DR) UHMWPE must show higher recovery stress. However, the degree of polymer chain elongation does not depend on the DR in straightforward manner. Orientation of UHMWPE at a temperature lower than melting point, is accompanied by gauche-trans transitions in chain segments in the amorphous phase, i.e. the dominant process is the straightening of macromolecules, while the orientation at temperatures in the vicinity of melting point of the polymer is dominated by interlamellar slip due to the change of the crystalline phase. Therefore, the orientation of UHMWPE fibers at the temperature of 110 °C, as performed in this study, leads to the dominance of gauche-trans transitions in the amorphous phase, which allows obtaining high recovery stresses at low drawing ratio.

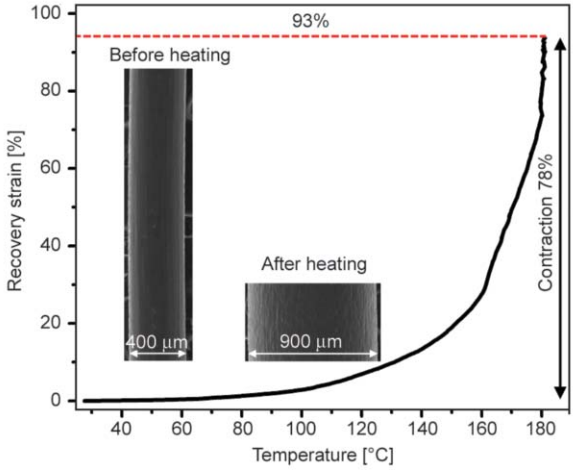


**Figure 4.** Recovery strain curve of UHMWPE fibers (predeformation strain of fibers is 500%).

Drawn UHMWPE fibers (at *DR* 6, i.e. pre-deformation strain 500%) have a recovery strain of 93% and can contract by 78% during heating, as shown in Figure 4. As suggested, the contraction of UHMWPE fibers occurs due to the entropic elasticity of the macromolecules, when the elongated macromolecules tend to occupy a more stable state with higher entropy value. In contrast to UHMWPE fibers, the reversible contraction of nylon fibers (4%) and polyethylene fibers (0.3%) is much lower, since it is governed by negative thermal expansion. Therefore, the high contraction of UHMWPE fibers enables obtaining artificial muscles with a high stroke.

The recovery strain of 93% has been achieved for the fibers manufactured from UHMWPE with specific structure (which macromolecules are able to slip along each other during drawing process) and high drawability. Such UHMWPE possesses less entanglement coupling, compared to other UHMWPE grades. Since one-way SME in UHMWPE occurs due to the high entanglement macromolecules coupling, which acts as a physical cross-linking [16], UHMWPE with high drawability partially loses the ability to return to the permanent shape during heating. Finally, we can conclude that the combination of particular UHMWPE grade and the applied drawing parameters lead to the creation of irreversible changes in the structure of polymer and acceptable portion (7%) of shape irreversibility.

Figure 5 shows DMA results of storage modulus and tan δ for UHMWPE fibers with *DR* 6 and coiled artificial muscles. The storage modulus of UHMWPE fibers with *DR* 6 decreases during heating, Figure 5a, which is a consequence of the increase in the viscous behavior of the material. The curve of a storage modulus has a bend around 75 °C, which can be correlated with α relaxation in UHMWPE. According to [23, 24], α relaxation is attributed to the chain motion in the crystalline phase. The tan δ (measure of energy dissipated) increased rapidly at around 130 °C, Figure 5a, increases rapidly at about 130 °C, Figure 5a, and this indicates the onset of melting in the drawn UHMWPE fibers. This is in good accordance with DSC data on the onset of UHMWPE fibers melting, as shown in Figure 2b. The tan δ demonstrates the beginning of rubbery region for UHMWPE fibers with *DR* 6.

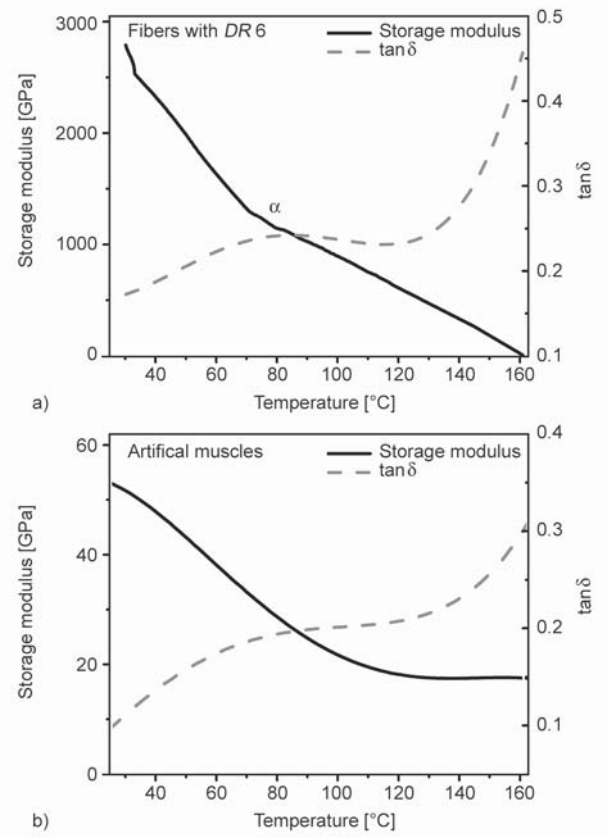


**Figure 5.** Storage modulus (solid line) and tan δ (dash line) for a) fibers with *DR* 6 and b) artificial muscles.

DMA results of storage modulus and tan δ measurement explain the behavior of recovery stress and recovery strain for UHWMPE fibers with SME. The increase of the temperature causes an increase in the molecular mobility in UHMWPE leading to an increase in the viscous behavior. During the recovery stress measuring, both tips of the UHWPE fiber were fixed in clamps so that the distance between the clamps, so the distance of clamps did not change during experiment. Therefore, any change in the volume of the material (in our case: a change in volume in the direction of the fiber axis) results in a linear increase in the recovery stress. The increase of the recovery stress of UHMWPE fiber relates with a practically linear drop of the storage modulus.

According to Figure 4, intensive changes in the recovery strain behavior begin to occur at the temperatures above 75 °C. This temperature can be considered as the activation temperature of the SME in the drawn UHWMPE fibers. According to the DMA data, this temperature corresponds to the α relaxation that is attributed to the chain motion in crystalline phase. The most intensive change of the recovery strain is observed at temperatures above 150 °C, which corresponds to complete melting of the crystalline

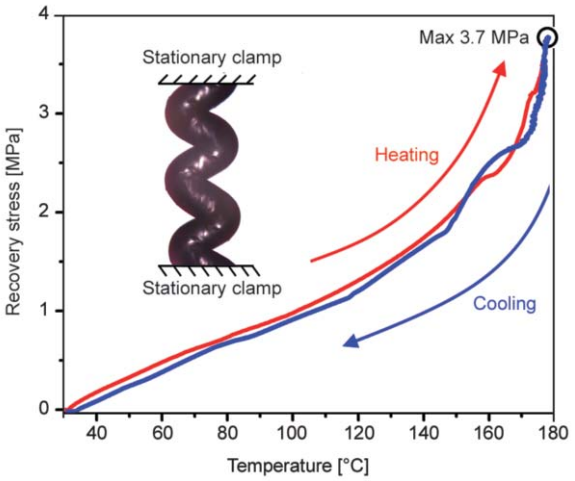


**Figure 6.** Isometric recovery stress of the artificial muscles based on coiled UHMWPE fibers.

phase and starting of the rubbery state for UHMWPE fibers. The crystalline phase in UHMWPE acts as a physical cross-linking of molecular chains [16, 25], acts as a skeleton, which keeps the deformation unless the crystalline phase breaks down during melting.

The obtained UHMWPE fibers as a basis for coiled artificial muscles have a number of advantages over other available commercial UHMWPE fibers:

– the ability to obtain fibers of almost any diameter. Commercial UHMWPE fibers produced by the gel-forming method have a diameter of 9 to 30 μm;
– higher recovery stress;
– a simpler and cheaper production technology.

Figure 6 shows the evolution of isometric recovery stress in the coiled UHMWPE artificial muscle. Gradual increase in the recovery stresses up to 1.7 MPa is observed while temperature increases up to 140 °C. In the temperature range from 140 to 180 °C, the slope of the curve however increases and recovery stress reaches up to 3.7 MPa. Coiled UHMWPE fibers show significant decrease in the recovery stresses, compared to those for drawn UHMWPE fibers, Figure 3. The storage modulus of artificial muscles is also significantly less than the storage modulus for drawn fibers, Figure 5. This effect can be related to the relaxation during annealing and essentially reduced stiffness of the coiled structure. During cooling the recovery stress of the artificial muscle gradually decreases. The hysteresis of the isometric recovery stress is not noticeable at heating-cooling cycling of the artificial muscle.

The maximum muscle stroke of the artificial muscles based on the twisted UHMWPE fibers with coil

index of 2.1 and constant stress of 1 MPa was measured as 64%, as shown in Figure 7. Video 1 (www.youtube.com/watch?v=vbCpWUWbOuQ&feature=youtu.be) demonstrates a stroke of artificial muscles during heating/cooling in air flow. Two characteristic stages can be distinguished for the artificial muscle stroke behavior during heating. The first stage (from 50 to 150 °C) shows gradual contraction up to 28%. The second stage (from 150 to 160 °C) reveals the sharp dramatic contraction up to 36%. The dramatic contraction occurs due to complete melting of the crystalline phase. After melting of the crystalline phase and transition to a rubbery state, the mobility of molecular chains increases, which leads to a dramatic increase in the reactive stresses of the artificial muscles made of UHMWPE fibers. Upon cooling, the artificial muscle relaxation and stretching begins at a temperature below 132 °C. In this regard, the obtained artificial muscles show large temperature stroke hysteresis of heating-cooling cycle. The reasons for the observed hysteresis are the difference in melting and crystallization temperatures. The melting point is the transition point to the rubbery region, after which the SME is fully activated. However, when the artificial muscle is cooling the rubbery region ends with the crystallization temperature. The crystallization temperature is lower than the melting point by 28 °C, as shown in Figure 2c. Only after passing through the crystallization temperature, the internal stresses in the artificial muscles decrease and the relaxation is observed. According to the DSC data, Figure 2c, the onset of crystallization of artificial muscles occurs at 119.7 °C, but the actual relaxation of the muscle occurs already at 132 °C, as shown in Figure 7.

The difference between the melting point and the crystallization temperature is the reason for the observed hysteresis in case of artificial muscles based on UHMWPE. The observed effect also confirms that the main nature of working artificial muscles is SME in UHMWPE. For the artificial muscles working on the basis of the negative linear expansion coefficient, hysteresis is a little or not observed [12]. Since the negative linear expansion coefficient has almost linear temperature dependence.

Theoretical calculations of the stroke of torsion and coiled actuators were presented in [12, 26]. It was shown that the negative thermal expansion results in a large stroke of the fiber actuators. Similarly, it can be assumed that the contraction of the drawn

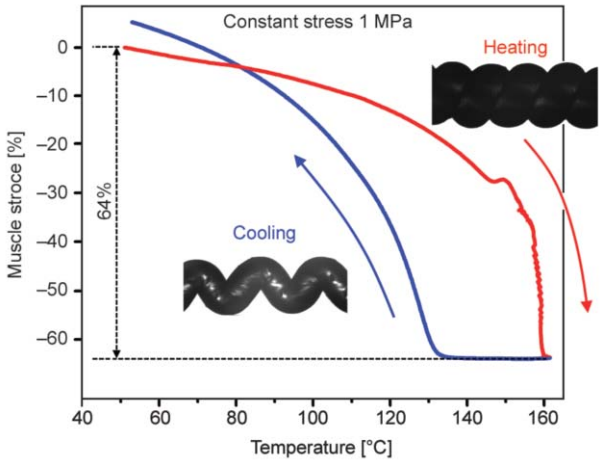


**Figure 7.** Contraction – relaxation cycle work of artificial muscle based on coiled UHMWPE fibers. The stress was 1 MPa; coil index was 2.1.

UHMWPE fibers also contributes to the large stroke of the coiled artificial muscles. However, the description of the relationship between the contraction of drawn fibers UHMWPE and the large stroke of coiled artificial muscles requires additional studies.

The resulting muscle stroke values are up till now the highest among the reported ones, for the artificial muscles based on twisted synthetic fibers with coil index of 2.1 at high temperatures. In [12], the coiled fibers were obtained using polyethylene fibers capable to contract for 16% during heating only up to 130 °C. The maximal contraction for 49% was achieved for nylon 6 coil artificial muscles with coil index of 5.5 at constant stress of 1 MPa [12] fabricated by wrapping of twisted fiber around a mandrel. In [26] actuators were obtained using linear low-density polyethylene fibers capable to contract for 23% at a temperature of 90 °C. Similar values for the maximal stroke were presented in [27], where the chemically cross-linked poly (ethylene-co-vinyl acetate) fibers with two-way shape memory effect were used for obtaining of coiled artificial muscles.

## 4. Conclusions

The possibility to fabricate artificial muscles based on coiled UHMWPE fibers was demonstrated. Previous studies considered the negative linear expansion in highly oriented amorphous-crystalline precursor fibers as the main driving force for thermal reversible contraction of coiled artificial muscles. In contrast, we have supposed another working principle of the artificial muscle – the entropic elasticity and one-way shape memory effect. In particular, the use of UHMWPE fibers capable to demonstrate

large contractions due to the entropic elasticity makes it possible to obtain coiled artificial muscles with a large stroke. The geometry characteristics (precursor fiber diameter, coil index and unloaded coil bias angle) seem to be less important than the functional properties of the precursor polymer fibers. Obtaining fibers with a high degree of contraction and small diameter is an important achievement, since high diameter of the artificial muscle complicates its heating and cooling, thereby reducing the response time.

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