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Abstract

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Disciplines

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Thermo-mechanical effects in the torsional actuation of twisted nylon 6 fiber

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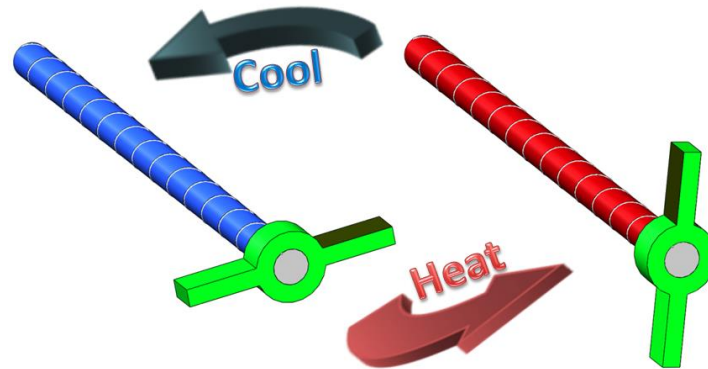
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Abstract

Thermally induced torsional and tensile actuators based on twisted polymeric fibers have opened new opportunities for the application of artificial muscles. These newly developed actuators show significant torsional deformations when subjected to temperature changes, and this torsional actuation is the defining mechanism for tensile actuation of twisted and coiled fibers. To date it has been found that these actuators require multiple heat/cool cycles (referred to as ‘training’ cycles) prior to obtaining a fully reversible actuation response. Herein, the effect of annealing conditions applied to twisted nylon 6 monofilament is investigated and it is shown that annealing at 200°C eliminates the need for the training cycles. Furthermore, the effect of an applied external torque on the torsional actuation is also investigated and torsional creep is shown to be affected by the temperature and load.

Keywords: torsional actuator, twisted polymer fiber, training cycles, annealing, torsional creep.

GRAPHICAL ABSTRACT



INTRODUCTION

Significant advancements in the performance of artificial muscles have been demonstrated in recent years by using twisted fibers and yarns made from a variety of materials such as carbon nanotubes, graphene, metallic nanowires and commercially-available polymer fibers¹⁻⁶. Volume increases induced by thermal, electrochemical or photonic stimuli or by absorption of small molecules causes partial untwist, or torsional actuation, in all of these systems. Very large torsional strokes of up to 250°/mm have been reported in electrochemically activated twisted carbon nanotube yarns³ and fast rotation speeds of up to 70,000 revolutions per minute have been recorded in twisted and coiled nylon 6,6 fiber when exposed to small temperature changes⁷. Potential applications of these actuators include microfluidic mixing³, microsensors⁸, photonic displays and energy harvesting devices^{7, 9}. Perhaps more importantly, this torsional mechanism can also be converted to large stroke linear (tensile)

actuation when the twisted fibers are formed into coils. Tensile strokes as high as 49% were reported for twisted and coiled nylon 6,6 fibers delivering power densities over 5.3 kW/kg with 2.48 kJ/kg contractile work capacity⁴ and greatly exceeding that produced by natural skeletal muscle (39 J/kg)^{10,11}. A major advantage of such actuators is that they can be easily fabricated from low-cost, commodity polymer fibers such as polyamides and polyesters used in fishing line and sewing thread.

One practical issue raised in early work on both twisted and coiled polymer fiber torsional and tensile actuators was the need for several ‘training cycles’ of heating and cooling to achieve fully reversible actuation^{4,12}. In the first few cycles, the degree of fiber untwist during heating exceeded the fiber re-twist that occurred during cooling^{12,13}. The magnitude of this effect has not previously been reported, nor has the molecular origin been described. Fiber twisting and coiling of oriented semicrystalline polymer fibers is an elasto-plastic mechanism where much of the inserted twist is lost due to elastic recovery unless the twisted fiber is thermally annealed while maintaining an external torque. Annealing or heat-setting is also commonly used to improve the mechanical properties of drawn polymer fibers by altering the fiber microstructure¹⁴⁻¹⁶. Here, we apply these annealing techniques to modify the structure of twisted nylon-6 fibers and investigate the effects on the torsional actuation behaviour.

In addition, the effect on its torsional actuation response of the applied external load acting on the fiber on its torsional actuation response is investigated. Tensile creep is known in drawn, semicrystalline polymer fibers and is likely to also occur during the thermally-induced torsional actuation of twisted polymer fibers. The magnitude of this effect has yet to be established, and here we investigate torsional creep in twisted nylon 6 fibers when subjected to small temperature changes.

EXPERIMENTAL

Sample fabrication

To fabricate the samples twist was inserted to the commercially available polymer fibers using a variable speed DC rotary motor ¹² while keeping the fibers well stressed at 10 MPa axial stress. 500 μm diameter monofilaments of nylon 6 (Sport Fisher monofilament fishing line) were twisted (~ 445 turns per meter) just before the first coil was due to form as a result of overtwisting (saturation). In this saturated state, the twist angle on the surface of fibers remained consistent ($\sim 35^\circ$ from the fiber axis) amongst all samples. Heat-setting of the twisted samples were conducted at acceptable temperatures which were held well below of the melting temperature of nylon 6. Differential scanning calorimetry (DSC) tests (TA Instrument DSC Q100) were conducted on as-received and twisted nylon 6 fibers, confirming their actual melting point. Heating rate was set at $10^\circ\text{C}/\text{min}$ and cooling rate was at $20^\circ\text{C}/\text{min}$ by using a 50 ml/min compressed nitrogen flow as the purging gas.

To study the effect of heat-setting on the actuation performance of twisted fibers, two different annealing approaches were conducted to permanently set the number of twists inserted and the helical structure introduced. In our previous studies, the heat-setting process used after the twist insertion was thoroughly described ¹². Following this process, four different samples of similarly twisted fibers were heat-set at 120, 150, 175, and 200°C for 2 hours followed by a 2 hours cooling period at room temperature (will be denoted ‘Set A’ hereafter). In the second approach (denoted ‘Set B’ hereafter), heat setting of twisted fibers was performed *in situ* and during the twist insertion process. Fig. 1 shows the schematic illustration of this simultaneous twisting/heat-setting process. A two-end open electric heating furnace was used through which the fiber was suspended. A programmable temperature controller (Electro Chemical Engineering Pty Ltd, Australia) was used to sense

and control the temperature throughout the furnace. Temperature was maintained at 120, 150, 175, and 200°C depending on the parameter chosen for fabrication process. Heating the sample during twist insertion causes a decrease in torsional stiffness of the fibre ¹²; hence, the number of twists required to form the first coil become higher compared to the twists needed in ambient temperature. Therefore, we inserted the same number of twists (~445 turns per meter) in ‘set B’ fibres as has been inserted in ‘Set A’ fibres. After twisting the fibers were then left for 2 hours to cool down at atmospheric conditions. The sample was then taken out from the furnace and was directly used for actuation tests.

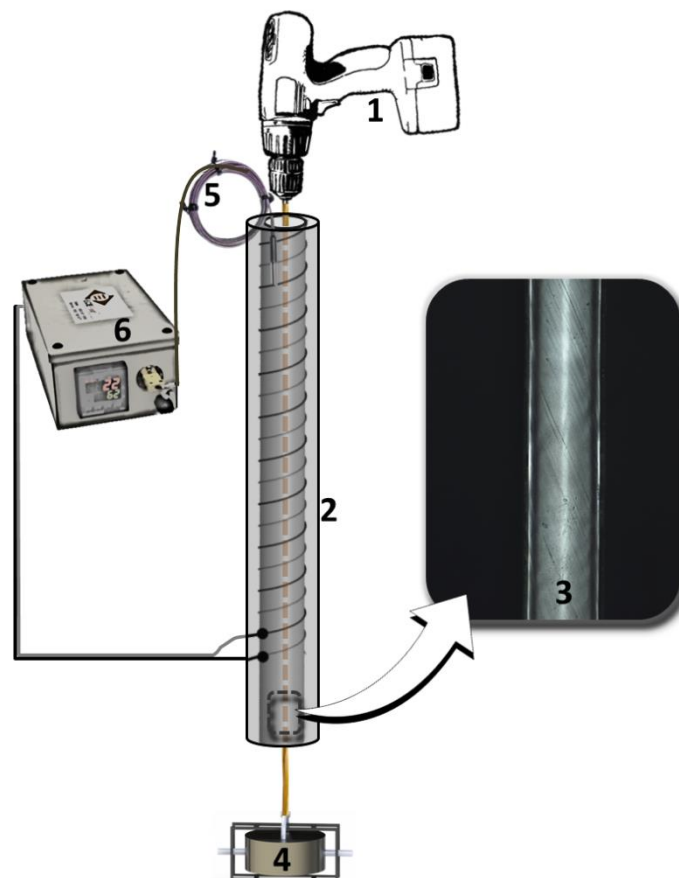


Figure 1. Experimental set up for simultaneous fiber twisting/heat-setting process: (1) DC rotary drill; (2) electrically driven heating furnace with proper insulation; (3) fiber under twist; (4) weight stone to apply stress to the fiber with rotation blocker to prevent untwist; (5)

thermocouple for sensing inside temperature of the furnace; (6) programmable temperature controller maintaining the required power delivered to the furnace.

Thermogravimetric (TGA) tests

To study the effect of water content on the actuation performance of twisted fibers exposed to moisture, thermal gravimetric analysis TGA (METTLER TOLEDO DSC/TGA) was employed. Initially, annealed samples were environmentally dried for ~24 hours and then, taken to the TGA chamber to be thermo-gravimetrically tested. The amount of water loss was analysed by heating the samples to 150°C. For comparison, other samples taken after annealing process were desiccator-dried (with anhydrous calcium chloride desiccant) for 24 hours to remove possible moisture. The dried samples were also tested with TGA and the results were compared with non-desiccated samples.

Torsional actuation tests

An in-house-built testing apparatus was used to perform thermally induced torsional actuation tests ¹². The test assembly was equipped with a DC power supply, programmable temperature controller and a dual mode lever arm force/distance transducer system (Aurora Scientific 305B). Twisted nylon 6 fibers were thermally actuated by electrical heating of the furnace. The fibers were fixed at one end and the other end was attached to a shaft supported by two near frictionless air bearings. Free rotation of actuated fibers was measured optically by placing an optical microscope (ISSCO-OPTEK) focused axially to the bearing shaft.

The rotation of the shaft caused by the isotonic (constant torque) torsional response of the twisted fibers was measured continuously from the lever arm that was connected to the shaft by a fiber held at constant tension. Different isotonic tests reported herein were conducted under a range of constant small external torques (68, 140, 280, and 560 $\mu\text{N}\cdot\text{m}$) applied by the lever arm and opposing the fiber untwisting. Applied torque values were chosen well below of the torsional yield stress at which the fiber starts to deform permanently to the twisting / untwisting direction. Unless otherwise stated, a constant twisted sample length (70 mm) and a constant temperature range (26-62°C) at average heating and cooling rates of 3.6°C/min and 1.8°C/min, respectively, were programmed and used for all the actuation tests.

RESULTS AND DISCUSSION

Differential scanning calorimetry

The mechanical and physical properties of semicrystalline polymers are strongly related to their microstructure, which is here investigated by DSC. Fig. 2(a) shows the DSC curves obtained from a 500 μm diameter as-received nylon 6 fiber when heated from 10°C to 300°C and then cooled. The melting temperature was $\sim 223^\circ\text{C}$ and the area of melting peak was 85.23 J/g from which a degree of crystallinity of 36% was calculated by comparison to the heat of fusion reported for a perfectly crystallized nylon 6 sample (239 J/g)^{17, 18}. Fig. 2(b) and 2(c) shows the DSC results for highly twisted fibers annealed at 120°C (Set A) and 120°C (Set B), respectively. Both materials show similar DSC thermograms with a slight increase in the melting temperature of 1-2°C and a small decrease in the degree of crystallinity by 1-2% when compared with the as-received fiber. The similar phenomenon was observed from the recrystallization peaks (on cooling) of different samples while recrystallization enthalpies decreased for twisted fibers compared to the as-received one.

Tsujimoto *et al.* and co-workers have previously investigated the effect of twist and annealing on the microstructure of nylon 6 monofilaments ¹⁹. They observed that twisting slightly decreases the degree of crystallinity by partial destruction of lamellae. The lamellae are initially oriented perpendicular to the fiber long axis and remain perpendicular to the twist angle for low to medium levels of twist insertion. Twist strain is accommodated by distortion of the amorphous regions and by re-orientation and partial destruction of the lamellae. It was further observed that annealing of the twisted fibers at 170°C for 15 minutes when held at constant length did not alter the degree of crystallinity, but stabilised the structure possibly due to better packing within the amorphous regions ¹⁹.

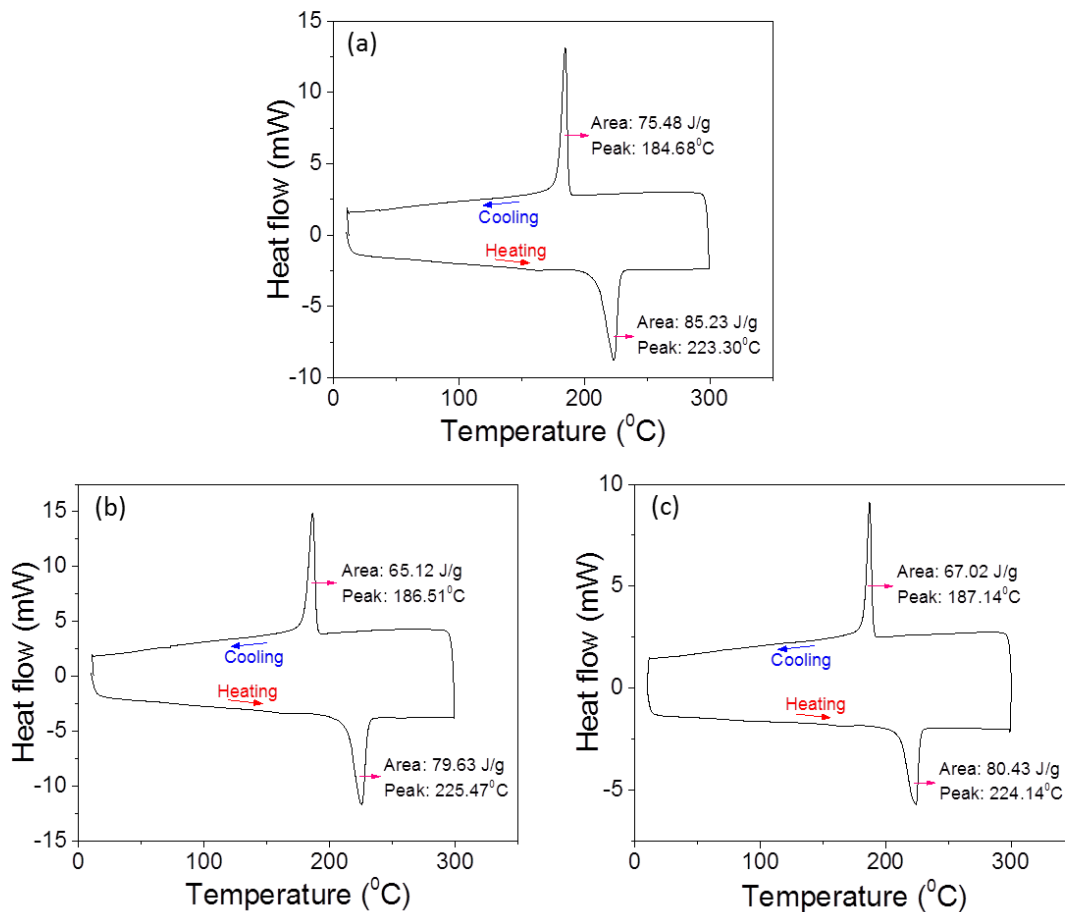


Figure 2. Differential scanning calorimetry result nylon 6: (a) as-received fiber; (b) highly twisted fiber annealed after twist insertion; (c) highly twisted fiber annealed during twist insertion.

Moisture content in the twisted samples

Due to the presence of hydrogen bonds, nylon 6 absorbs considerable amount of water when left in natural condition^{20, 21}. In particular, amorphous phase water sorption of nylon 6 is largely recognized²¹⁻²³ which acts as a plasticiser to reduce the glass transition temperature²⁴. This process may influence the actuation performance, since the thermal expansion coefficient of polymers increases above T_g and thermal expansion drives the torsional actuation in twisted polymer fibers. TGA was performed to evaluate the amount of water present in the twisted fibers. Fig. 3 shows the TGA results for the twisted and annealed (200°C) nylon 6 samples when heated from 30°C to 150°C. Fig. 3(a) shows gradual but continuous weight loss of up to 2.7 wt% for the non-desiccated sample, whereas the desiccated sample had a significantly lower water loss of 0.8 wt% when heated over the same temperature range [Fig. 3(b)]. In both cases, successive heating cycles have shown almost negligible sample weight changes in that particular temperature region. These results indicate that the non-desiccated sample retained a small amount of atmospheric water. The reduction in mass due to water loss for both samples during their first heating cycle over the temperature range of 30-62°C was small. Since torsional actuation tests were conducted over

this temperature range, it was concluded that the effect of moisture loss during actuation tests could be ignored in this study.

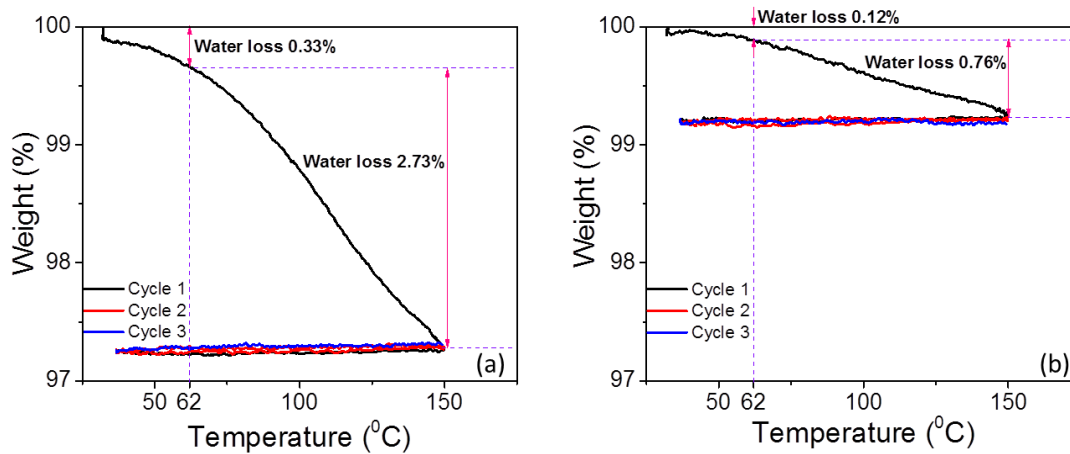


Figure 3. Thermogravimetric test results of twisted nylon 6 fiber: (a) as-twisted non-desiccated sample; (b) 24 hours desiccator dried sample.

Effect of annealing conditions on torsional actuation

Fig. 4 shows the torsional stroke recorded during the first heat/cool cycle for twisted fiber samples prepared by both annealing methods. Almost identical torsional strokes were obtained for samples prepared by both methods when the annealing temperature was consistent. Regardless of the annealing method used, those fibers annealed at 120°C and 150°C showed large untwisting rotations of up to $-7^{\circ}/\text{mm}$ during heating, but with a smaller re-twisting recovery during cooling ($+2^{\circ}/\text{mm}$). As the annealing temperature increased the fibers exhibited less untwisting on first heating but with considerably higher degrees of reversibility. Samples annealed at 200°C provided the smallest rotation of $-1.7^{\circ}/\text{mm}$ on heating and almost the same re-twisting on cooling. This result is the same as reported previously¹² for identical 500 μm diameter nylon 6 twisted fiber that had been annealed at

120°C but had been further ‘trained’ for several heat / cool cycles to achieve full torsional reversibility.

The effect of annealing temperature on the reversibility of torsional actuation of these twisted nylon 6 fibers may be understood from previous studies of heat-treatment induced microstructural changes in axially drawn nylon 6 fibers. Prior work has shown that important microstructural differences occur when fibers were annealed under tension or when slack²⁵. Considerable shrinkage occurs in the length direction in slack fibers as a result of molecular refolding processes, but these mechanisms are significantly inhibited by applied tension. In the present study, annealing was conducted at constant fiber length thereby potentially limiting molecular reorganization. First heating during torsional actuation testing of incompletely annealed twisted fibers may allow sufficient molecular mobility for some reorganization to occur. Shrinkage along in the chain orientation direction, which forms helically paths in these twisted fibers, would then promote fiber untwisting. The result is a large untwisting during first heating due to both irreversible molecular reorganization and reversible thermal expansion effects. Twisted fiber samples annealed at 200°C appear to be immune from these irreversible processes, suggesting that the higher annealing temperature allows a stable molecular structure to form even when the fiber was held at constant length during heat treatment. Indeed, Tsujimoto *et al.* have shown that annealing of twisted nylon 6 fibers at 170°C at constant length stabilises the molecular structure, possibly by increased molecular packing in amorphous regions¹⁹.

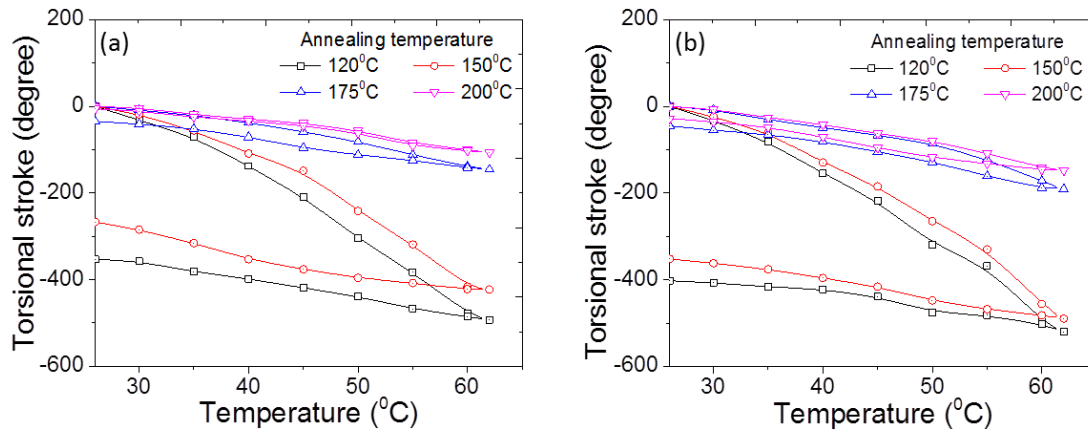


Figure 4. Torsional actuation test results of 70 mm long twisted nylon 6 fiber samples prepared at different annealing temperatures: (a) samples annealed after twisting (Set A); (b) samples annealed during twisting (Set B). (Each heating process is shown to start at ‘0’ torsional stroke for all cases).

Effect of external torque on torsional actuation

The effect of an external torque applied in the twist direction on the torsional stroke during the first heating and cooling cycle was next investigated for samples prepared by annealing at 200°C (Fig. 5). With increasing applied external torque, the untwisting rotation on heating diminished in magnitude and the retwisting on cooling increased. The net result was an increase in sample twist after the first heat / cool cycle for samples tested at high external torque. The effect of the applied torque was particularly noticeable for samples that were annealed during twisting and was smaller for samples that were annealed post twisting. It was found that two heat / cool ‘training cycles’ were required at a fixed external torque before the sample showed equivalent untwist / retwist magnitudes during heating and cooling. Fiber samples tested at the maximum torque for three heat / cool cycles were then re-tested at the minimum torque. The first heat / cool cycle at the low torque condition showed a reversibility of 80-95% with some unrecovered untwist.

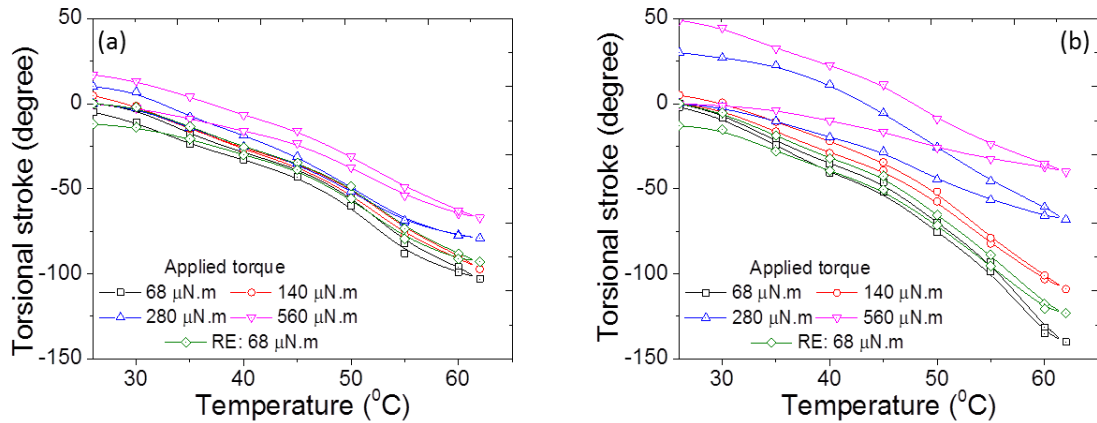


Figure 5. Torsional actuation test results of 70 mm long twisted nylon 6 fiber samples tested with different externally applied torque. (a) samples annealed after twisting (Set A); (b) samples annealed during twisting (Set B).

Torsional creep tests were conducted to further investigate the effect of applied torque on the torsional actuation response. Longer time exposure to an external torque applied in the twist direction resulted in a gradual increase in fiber twist with the effect more pronounced at higher torques and higher temperatures (Fig. 6). The observed increase in twist due to this torsional creep correlates approximately with the results of Fig. 5 when the temperature and timescale of the heat / cool cycles are considered. In addition, the sample tested initially at the maximum torque and then re-tested at the minimum torque showed some recovery of the torsional creep. Again, this observation is in qualitative agreement with the assumption that there was some net untwisting during the heating / cooling of the sample tested at the minimum torque immediately following testing at the maximum torque.

Tensile creep is well documented in polymers, and studies of nylon 6,6 demonstrate appreciable increases in compliance, even at room temperature and at the timescales of the experiments described here ^{26, 27}. This torsional creep can be explained as gradual deformation of oriented polymer chains. Thermal annealing treatment has been shown to aid

creep deceleration in semicrystalline polyester and polyamide materials²⁸. The reduced creep rate was reportedly due to the formation of undeveloped crystallites in the amorphous phase during annealing which serve as obstacles for sliding of chain ends between polymer chains²⁸. In the present study, however, the high temperature annealing was unable to completely eliminate the torsional creep process when high external torques were involved. Modelling approaches, such as proposed recently by Fancey²⁹, may allow the prediction of time-dependent torsional strokes at different loads and temperatures. Preferably, the creep in torsional actuators should be negligible and techniques used to reduce tensile creep in polymers, such as fiber reinforcement, or crosslinking are worthy of further investigation.

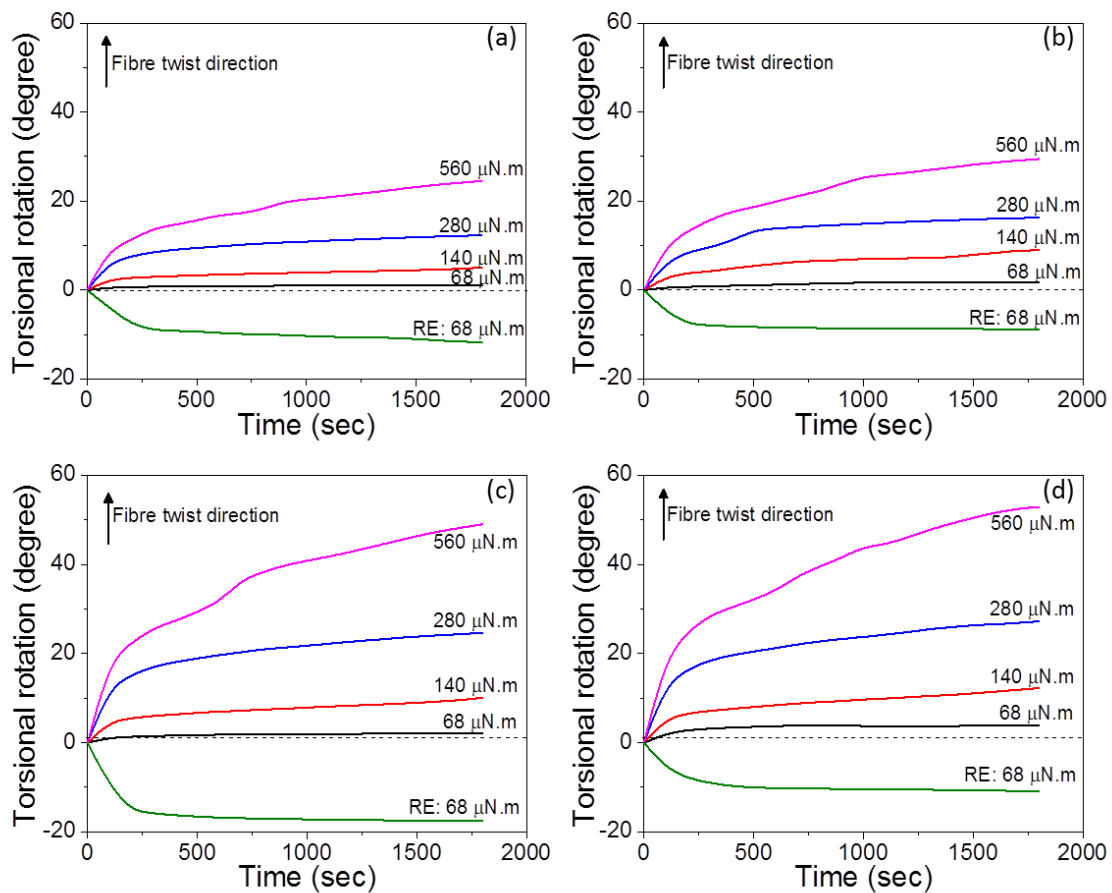


Figure 6. Time based torsional creep of twisted nylon 6 fibers under different isotonic torques. Tests were conducted at 26°C: (a) annealed at 200°C after twisting (Set A), (b)

annealed at 200°C *in situ* twisting (Set B). Tests conducted at 62°C: (c) annealed at 200°C after twisting (Set A), (d) annealed at 200°C *in situ* twisting (Set B).

CONCLUSIONS

The purpose of this study was to further explore the torsional actuation of twisted nylon 6 monofilament and to optimise their fabrication conditions to achieve a more stable actuation performance under varying isotonic torques. Nylon 6 monofilaments were twisted and annealed at several temperatures below their melting point. One set of samples was prepared by heat-setting during twisting, whereas another sample set was annealed after twist insertion. Heating and cooling of the samples over a 36°C range resulted in cyclic torsional stroke due to untwisting (heating) and retwisting (cooling) of the fibers. As in prior studies, samples annealed at low temperature (< 175°C) showed a larger untwisting and smaller retwisting during the first heat / cool cycle. In contrast, samples annealed at 200°C showed complete reversibility in the first cycle, without the need for any ‘training’. The application of an external torque caused torsional creep that was accelerated by increases in temperature and torque. The baseline twist in the fiber readjusts to a new level during the first two heat/cool cycles when the external torque is changed. Thereafter, the torsional untwist and retwist remain consistent in subsequent heat / cool cycles at constant torque. Heat setting can, therefore, be used as an effective means for fixing a consistent torsional actuation response.

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