

Silver-Nanowires Coated Pitch-Tuned Coiled Polymer Actuator for Large Contractile Strain under Light-Loading

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We report a pitch-tuned coiled polymer actuator (CPA) with a compliant silver-nanowires (AgNWs) electrode. For a pitch-tuning of the CPA, we sequentially implement a twist-insertion for coiling a nylon-6 fiber, mechanical pre-stretching, and a couple of thermal treatments. When heated under a light-loading, the pitch-tuned CPA can produce a contractile strain far beyond the original one with a limited space to contract. Since the silver-nanowires electrode established on the coiled structure retains a fairly consistent electrical property allowing repetitive Joule-heating under a large uniaxial elongation with loading, the synergetic benefits from pitch-tuning and adopting the AgNWs as a Joule-heating electrode enables the CPA to produce electrically-controllable and large contractile strain under a light-loading that can be utilized for an artificial muscle of soft micro-robot.

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1. Introduction

In last decade, many studies on artificial muscles for use in soft robots and wearable devices have been introduced. As a wearable artificial muscle, development of a power-assist exoskeleton opens an opportunity to improve functionality of human muscle,¹⁻³ while there are difficulties in achieving precise stroke-control required for implementing complex tasks that human do and heavy weight as well as rigidity of frame still make humans uncomfortable for wearing. As an alternative, soft actuators with a light-weight and flexible nature have given much attention due to their feasibility for miniaturization as well as increased demands for soft robots.⁴ A number of soft actuators have been developed by using smart materials such as dielectric elastomers, conductive polymers, ionic polymer-metal composite (IPMC), shape memory alloys (SMAs).⁵⁻⁹ Although these approaches have their own benefits, there is a technical challenges in securing large output force with rapid response comparable to that of human muscle.

Recently, as a new class of artificial muscle, a soft actuator based on coiled nylon fiber has been proposed by Haines.¹⁰ The coiled polymers actuator (CPA) shows a great potential for artificial muscle due to its outstanding thermally-induced contractile strain, low cost fabrication, light-weight structure, and high mechanical strength. In order to secure operational stability of the coiled actuator, many researchers have

suggested not only fabrication methodologies to optimize twist-insertion, annealing, and thermally-induced training processes, but also various coiling materials.¹⁰⁻¹² The efforts remarkably contribute to improving thermally-induced contraction capability and to drawing feasible applications, but most of researches have been mainly focused on improving performance under heavy-loading.

Meanwhile, researches to find an effective driving force securing stable thermo-mechanical performance of the coiled polymers has also been widely studied by implementing chemical, hydronical, photonical, and electrical heating¹⁰ as well as ambient temperature change.^{13,14} Particularly, as a direct heating approach, the electrical heating by using conductors such as carbon nanotube (CNT) sheet, a steel wire,¹⁰ metallic particles,^{12,15} conductive elastomer¹⁶ contributes to rapidly and comfortably producing thermally-induced contractile strain without using external heating source that can usually be an obstacle for miniaturization. The electro-thermal operation allows practical applications of the coiled actuator for active-textiles¹¹ as well as robotic systems such as a wearable garment¹⁶ and a robotic hand.^{17,18}

Here, we propose a pitch-tuned CPA with a compliant Joule-heating electrode using silver-nanowires (AgNWs). The pitch of the coiled polymer prepared via twist insertion of a nylon-6 fiber is modulated by uniaxial pre-stretching and a couple of thermal treatments in sequence. The pitch-tuning contributes to remarkably improving

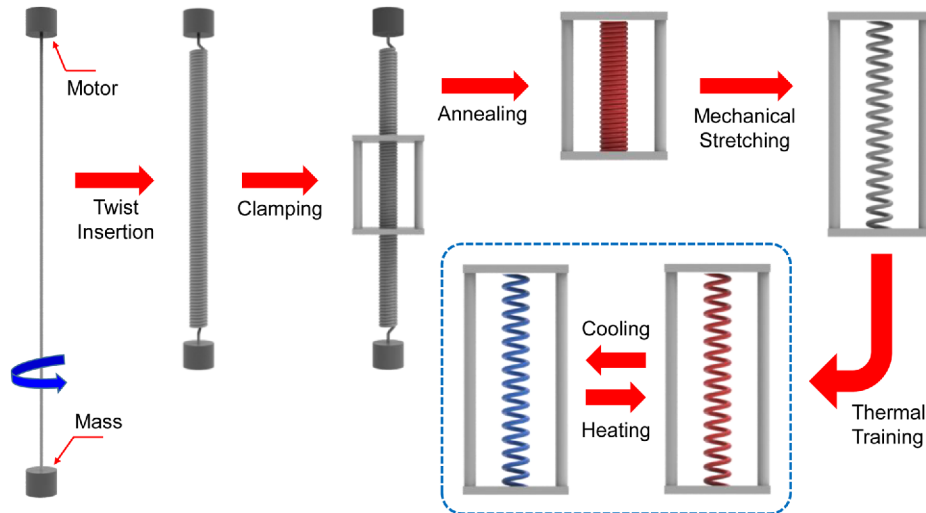


Fig. 1 An illustrated schematic preparation process for pitch-tuned coiled polymer

thermally-induced contractile strain of an original coiled-polymer under a light loading. For electrical operation of the coiled-polymer, AgNWs are adopted as a flexible conductor for Joule-heating electrode. A thin AgNWs layer constructed on the coiled structure exhibits a fairly reversible electrical property during repetitive loading-induced elongation and Joule heating tests. A combination of pitch-tuning and a highly compliant Joule-heating electrode enables demonstration of a CPA allowing reversibly-controllable large contractile actuation under a light-loading, which can be required for artificial muscle of soft micro-robots.

2. Experimental

2.1 Fabrication process

2.1.1 Pitch-tuned coiled polymer

We use nylon-6 monofilament fishing line (diameter: 285 μm), which is purchased from Torrey (Tournament SE), as a precursor fiber. Using the fishing fiber, a coiled actuator was fabricated by implementing four steps in sequence, which is illustrated in Fig. 1. In the first step, a twist was inserted into the precursor fibers (length: 1200 mm) with a twisting speed of 700 RPM under a load of 40 MPa. As the result of the twist insertion step, a coiled polymer with a length of 300 mm and a diameter of 365 μm (spring index of 1.3) was obtained. In the second step, the coiled-polymer clamped at a rigid fixture was thermally annealed in a vacuum oven at 170°C for 100 minutes. The third step is to adjust the pitch of the coil. After primary annealing process, the coiled-polymers were individually stretched with different percentage elongation of 10, 30, 50 and 70%, respectively, and then each coiled-polymer was thermally treated by continuously implementing 10 repetitive heating (at 175°C for 3 min)/cooling (for 3 min) processes in a temperature controllable chamber. Finally, in order to obtain pitch-tuned coiled-polymers with a consistently controllable-pitch, a training process was additionally implemented through repetitive heating/cooling processes with a constant ramp rate of 3°C/min in the temperature ranging from room condition to 175°C. Pitch-

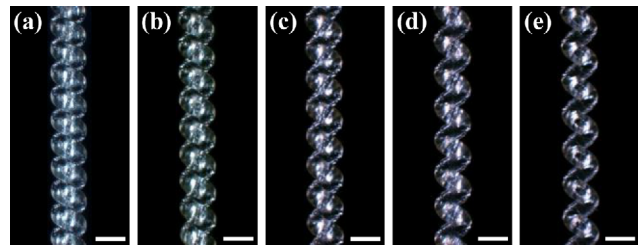


Fig. 2 Optical microscope images of the coiled-polymers with different pitches of (a) 0%, (b) 10%, (c) 30%, (d) 50%, and (e) 70%. The scale bar is 500 μm

tuning induced geometrical change of the coiled polymers was observed by using an optical microscope as shown in Fig. 2.

2.1.2 Compliant electrode coatings

Silver-nanowires is employed as a Joule-heating electrode for the pitch-tuned CPA. An AgNWs solution, which is a dispersion of AgNWs (average diameter: 25 \pm 5 nm, average length: 35 \pm 5 μm) in isopropyl alcohol (IPA) with a concentration of 0.5 wt%, is purchased from Nanopyxis. By adopting dip-coating and spray-coating methodology individually, a thin AgNWs layer is formed on a surface of the pitch-tuned coiled polymer. In advance of the AgNWs coating, both ends of the coiled polymer are clamped at a couple of rigid frames as maintaining a tension preventing the coiled polymer from being undesirably buckled. For dip-coating, the clamped coiled polymer is soaked into a cylinder filled with an AgNWs solution during 20 seconds, taken out of the cylinder, and dried in room condition in sequence. For spray coating, the clamped coiled polymer is attached on a zig. The AgNWs solution was sprayed on the coiled polymer as continuously rotating the zig. The AgNWs layer formed on the coiled polymer was dried at room temperature.

2.2 Characterization

Thermo-mechanical behavior of the pitch-tuned coiled polymers

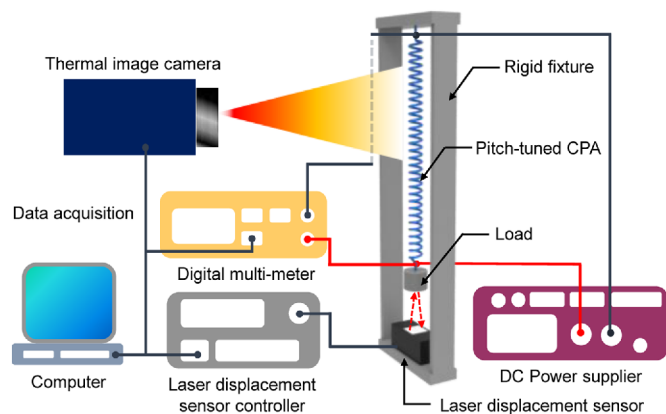


Fig. 3 An illustrated schematic performance test system for the CPAs

was evaluated by using a thermomechanical analyzer (RSA-G2, TA Instruments). For the testing, both ends of the pitch-tuned coiled polymer were clamped at the rigid fixture and pre-loaded with 1N. As maintaining the loading condition, temperature dependent contractile deformation was monitored by cyclically changing temperature in the range of room temperature and 170°C in a chamber with a consistent heating and cooling rate (3°C/min). In order to study electrical stability of the AgNWs electrode on the pitch-tuned coiled under mechanical elongation with loading, electrical resistance change responding to repetitive mechanical stretching/relaxation was measured by using a digit multi-meter (34465A, Keysight Technologies).

2.3 Performance test

Joule-heating induced actuation performance of the pitch-tuned CPA with an AgNWs compliant electrode was evaluated by using a performance test system, which is illustrated in Fig. 3. Input current for Joule-heating was modulated by using a DC power supply (2220-30-1, Keithley). During Joule-heating, temperature dependent contractile strain of the CPA was measured by a laser displacement sensor (LK-H055, KEYENCE) as monitoring resistance of the AgNWs electrode and temperature on the surface of CPA through a digit multi-meter and a thermal image camera (A65sc, FLIR), respectively.

3. Results and Discussion

3.1 Thermo-mechanical behavior of the pitch-tuned coiled polymers

Pitch-tuned coiled-polymers were prepared by sequentially implementing three steps composed of mechanical pre-stretching in uniaxial direction, annealing, and cyclic training process. After annealing process, we observed the modulated pitch tends to be irregularly changed when a load applied for the mechanical elongation is eliminated from the coiled-polymers. Since the structural uncertainty can cause unstable thermally-induced contraction/recovery behavior of the coiled polymers, we additionally adopted the cyclic training process that performs several cyclic heating/cooling steps continuatively. Although the tuned-pitch still becomes reduced to be less than desired one even after the training process, it contributes to remarkably

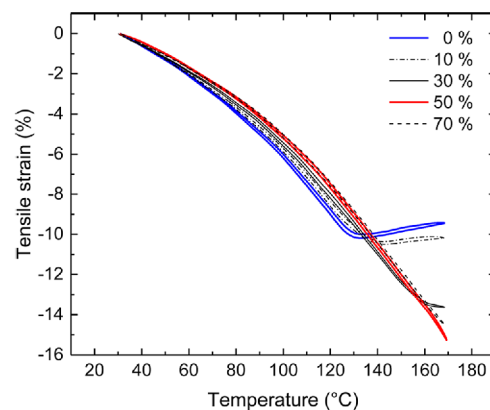


Fig. 4 Comparison of thermally-induced contractile strain profile for the original coiled-polymer and the pitch-tuned coiled-polymers with different pitches

reducing deviation of the tuned-pitch as securing fairly consistent reduction ratio. We note that for pitch-design of the coiled-polymer, an inevitable pitch-reduction, which is required to relieve thermo-mechanically-induced residual stress accumulating in the coiled structure during the training process, should be considered.

To investigate the influence of the pitch-tuning on thermally-induced contraction behavior of the coiled-polymers, their contractile strain profiles with temperature under a light loading, 1 N were measured during a cyclic operation with a consistent heating/cooling rate, 3°C/min. As shown in Fig. 4, regardless of the pitch-tuning, when heated the coiled-polymers intrinsically produce thermally-induced contractile strain and exhibit fairly well recovery from the deformed state as cooling down to room temperature. However, there is a significant difference in contractile strain profile as temperature becomes higher than 120°C. The pitch-tuned coiled polymer can produce more contractile strain, while the original one becomes stationary although temperature increases to 170°C. Particularly, in the case of the pitch-tuned coiled polymers, there is an increasing tendency of the contractile strain as percentage elongation for the pitch-tuning increases from 10% to 50% under 1N loading, suggesting that a contraction limit of the coiled-polymer can be modulated by intentionally forming a pitch. Due to the benefits from the pitch-tuning treatment, the coiled-polymer is capable of producing 50% larger strain than the original one under the loading of 1 N.

3.2 Compliant Joule-heating electrode for the pitch-tuned CPA

AgNWs are adopted as a joule-heating electrode suitable for the pitch-tuned CPA since the AgNWs are flexible, highly conductive, and less restrictive regarding formable geometry. By individually implementing dip-coating and spray-coating methodology, a thin AgNWs layer is established onto a surface of the coiled structure. Fig. 5 shows microscopic images of the AgNWs established on the coiled polymer via the coating methodologies. In the case of dip-coating, the AgNWs with a conductive network are formed on the coiled structure in a short time while the network structure becomes partially unstable due to formation of AgNWs solution droplets on the coiled structure, which results in irregular and locally agglomerated AgNWs during

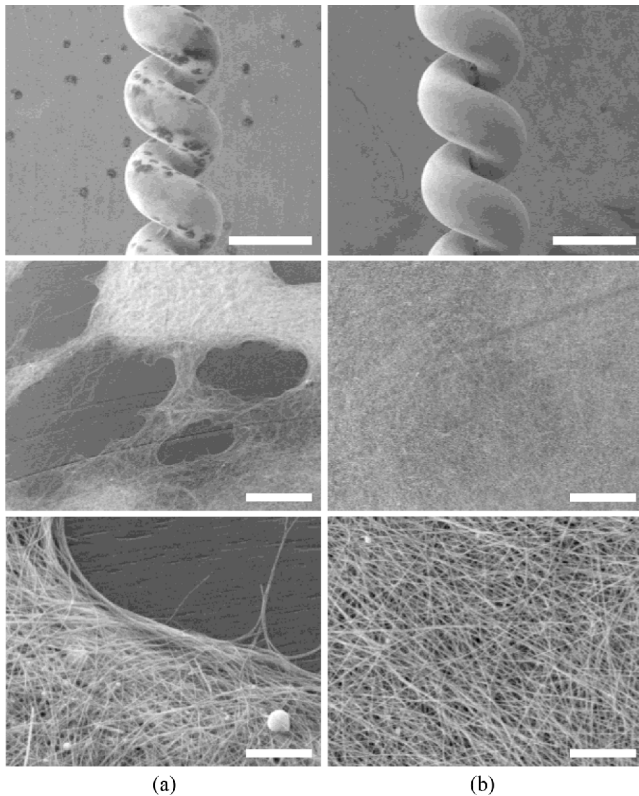


Fig. 5 SEM images of the AgNWs electrodes established on the surface of a pitch-tuned CPA by using (a) dip-coating and (b) spray-coating methodology. The scale bars are 500, 20, and 2 μm from top to bottom in order

drying process as shown in Fig. 5(a). On the other hand, the spray-coated AgNWs exhibits uniform and dense network formation on the coiled geometry, as shown in Fig. 5(b). It suggests that the coating methodology, which sprays out in a fine mists of the AgNWs dispersed in a highly volatile solvent, can contribute to reducing locally-agglomerated AgNWs.

Fig. 6 shows electrical resistance profiles of the AgNWs electrodes during repetitive elongation/relaxation cycles. The dip-coated AgNWs electrode shows a sharp increasing tendency in the resistance change with respect to the R_0 ($\Delta R/R_0$) as a loading-induced-strain increases as high as 50% although it can be fairly reversibly recovered to an initial resistance. In contrast, for the spray-coated AgNWs electrode, under the same cyclic loading condition the $\Delta R/R_0$ becomes much smaller, which is less than 5% as securing reversible recovery of the resistance. Even under heavier loading as large as 4 N, the $\Delta R/R_0$ only increases to 10% due to the structural benefits forming flexible and dense conductive pathways on the surface of coiled polymer. Based on the electrical stability against loading-induced strain, we adopted AgNWs forming via spray-coating as a compliant Joule-heating electrode for a pitch-tuned CPA.

Finally, Joule-heating performance of the AgNWs electrode is investigated for demonstration of electrically-controllable pitch-tuned CPA. As shown in Fig. 7, when an input current for Joule-heating increases to 32 mA, the temperature on the pitch-tuned CPA shows a

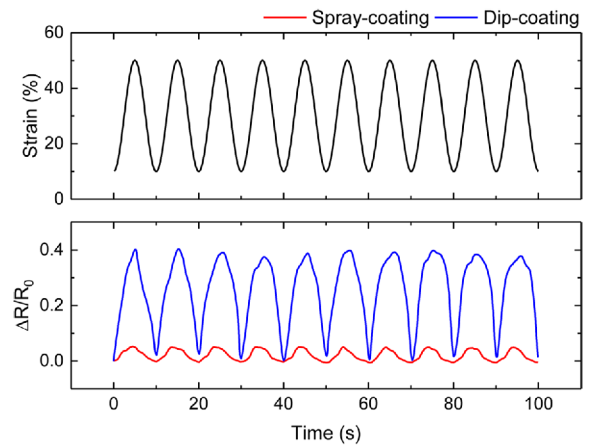


Fig. 6 $\Delta R/R_0$ change of the AgNW electrodes established via different coating methodology during 10 cyclic loadings of strain (continuous strain-relaxation cycles between 0 and 50% peak strain)

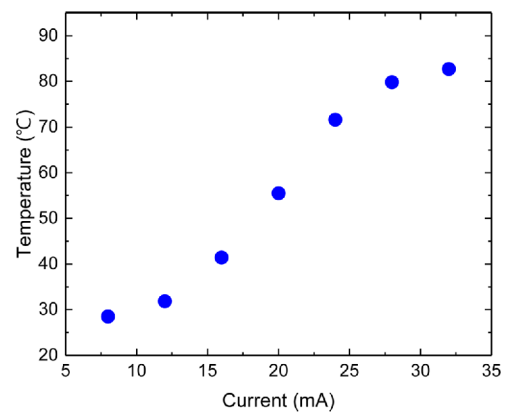


Fig. 7 An input current dependent Joule-heating temperature profile on the surface of the CPA

clear increasing tendency and then saturates to around 80°C, indicating that contractile strain of the pitch-tuned CPA can be intentionally modulated by controlling the input current. As shown in Fig. 8, when an electric current is applied to the AgNWs electrode, whole surface area of the pitch-tuned CPA is heated up simultaneously within 10 seconds and it is also cooled down in a short period of time after electric signal becomes off-state.

Particularly, the AgNWs electrode based electrically-controllable Joule heating allows the pitch-tuned CPA to programmably modulate the contractile strain as high as 15% even under much lower temperature than indirect heating with an external heating source, which is adopted for the study of the thermally-induced contraction behavior with pitch. It suggests that direct heating can be more effective approach to remarkably reduce an electrical power required for operation of thermally-driven actuator. Based on the study, we demonstrate a pitch-tuned CPA with the AgNWs electrode is capable of producing electrically-controllable, large and reversible contractile strain under a light-loading.

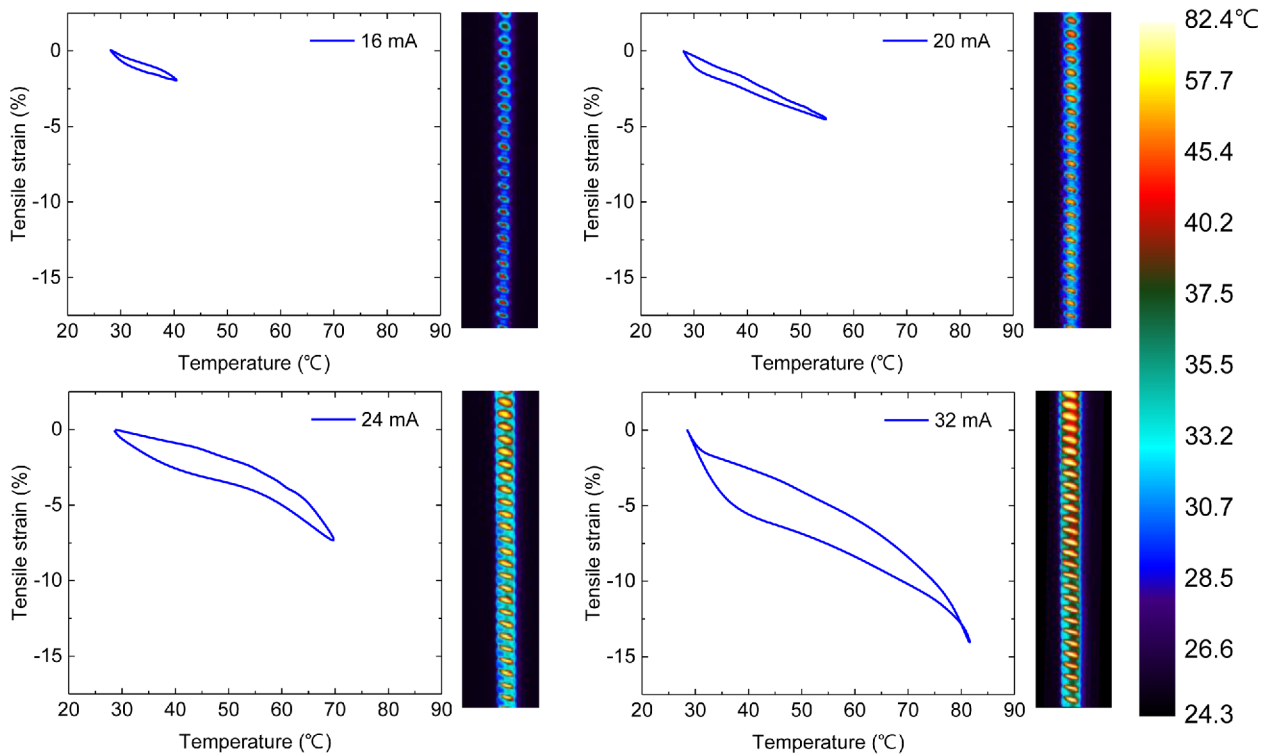


Fig. 8 Thermally-induced actuation performance and thermal image of a pitch-tuned CPA with a pitch of 50% responding to Joule-heating with different input currents of a) 16 mA, b) 20 mA, c) 24 mA, and d) 32 mA

4. Conclusions

In this study, we report a compliant AgNWs electrode based pitch-tuned CPA that is capable of producing a programmably-controllable and large contractile-strain under light-loading. The pitch-tuned CPA is prepared by sequentially implementing a twist insertion for coiling a nylon-6 fiber, pre-stretching, and a couple of thermal treatments. Based on parameter study for a pitch-tuning process, we established an effective way to control geometric structure of the coiled polymer actuator and to stabilize the geometric change. The pitch-tuning could lead to remarkable increase in contractile strain under a light loading of 1N. For demonstration of electrically-controllable contraction of a pitch-tuned coiled polymer, AgNWs is studied for a compliant Joule-heating electrode. The AgNWs electrode with a uniform network formation on the coiled structure is highly conductive, robust against repetitive mechanical elongations with loading, and also retains a fairly reversible electrical property during repetitive Joule-heating tests. The synergetic benefits from pitch-tuning and adopting the AgNWs as Joule heating electrode enables the CPA to produce electrically-controllable, large and reversible contractile strain under a light-loading.

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REFERENCES

- Zhang, J., Fiers, P., Witte, K. A., Jackson, R. W., Poggensee, K. L., et al., "Human-in-the-Loop Optimization of Exoskeleton Assistance during Walking," *Science*, Vol. 356, No. 6344, pp. 1280-1284, 2017.
- Heo, P., Gu, G. M., Lee, S.-J., Rhee, K., and Kim, J., "Current Hand Exoskeleton Technologies for Rehabilitation and Assistive Engineering," *International Journal of Precision Engineering and Manufacturing*, Vol. 13, No. 5, pp. 807-824, 2012.
- Ko, C.-Y., Ko, J., Kim, H. J., and Lim, D., "New Wearable Exoskeleton for Gait Rehabilitation Assistance Integrated with Mobility System," *International Journal of Precision Engineering and Manufacturing*, Vol. 17, No. 7, pp. 957-964, 2016.
- Cho, K.-J., Koh, J.-S., Kim, S., Chu, W.-S., Hong, Y., and Ahn, S.-H., "Review of Manufacturing Processes for Soft Biomimetic Robots," *International Journal of Precision Engineering and Manufacturing*, Vol. 10, No. 3, pp. 171-181, 2009.
- Binh, P.C., Nam, D. N. C., and Ahn, K. K., "Modeling and Experimental Investigation on Dielectric Electro-Active Polymer Generator," *International Journal of Precision Engineering and Manufacturing*, Vol. 16, No. 5, pp. 945-955, 2015.
- Jeon, J.-H. and Oh, I.-K., "Introduction to Ionic Polymer-Metal Composite Actuators and their Applications," *Journal of the Korean Society for Precision Engineering*, Vol. 28, No. 11, pp. 1242-1250, 2011.

7. Bhandari, B., Lee, G.-Y., and Ahn, S.-H., "A Review on IPMC Material as Actuators and Sensors: Fabrications, Characteristics and Applications," *International Journal of Precision Engineering and Manufacturing*, Vol. 13, No. 1, pp. 141-163, 2012.
8. Lendlein, A. and Langer, R., "Biodegradable, Elastic Shape-Memory Polymers for Potential Biomedical Applications," *Science*, Vol. 296, No. 5573, pp. 1673-1676, 2002.
9. Lendlein, A., Jiang, H., Jünger, O., and Langer, R., "Light-Induced Shape-Memory Polymers," *Nature*, Vol. 434, No. 7035, pp. 879-882, 2005.
10. Haines, C. S., Lima, M. D., Li, N., Spinks, G. M., Foroughi, J., et al., "Artificial Muscles from Fishing Line and Sewing Thread," *Science*, Vol. 343, No. 6173, pp. 868-872, 2014.
11. Haines, C. S., Li, N., Spinks, G. M., Aliev, A. E., Di, J., and Baughman, R. H., "New Twist on Artificial Muscles," *Proceedings of the National Academy of Sciences*, Vol. 113, No. 42, pp. 11709-11716, 2016.
12. Park, J., Yoo, J. W., Seo, H. W., Lee, Y., Suhr, J., et al., "Electrically Controllable Twisted-Coiled Artificial Muscle Actuators Using Surface-Modified Polyester Fibers," *Smart Materials and Structures*, Vol. 26, No. 3, Paper No. 035048, 2017.
13. Mendes, S. S. and Nunes, L. C. S., "Experimental Approach to Investigate the Constrained Recovery Behavior of Coiled Monofilament Polymer Fibers," *Smart Materials and Structures*, Vol. 26, No. 11, Paper No. 115031, 2017.
14. Cherubini, A., Moretti, G., Vertechy, R., and Fontana, M., "Experimental Characterization of Thermally-Activated Artificial Muscles Based on Coiled Nylon Fishing Lines," *AIP Advances*, Vol. 5, No. 6, Paper No. 067158, 2015.
15. Mirvakili, S. M., Ravandi, A. R., Hunter, I. W., Haines, C. S., Li, N., et al., "Simple and Strong: Twisted Silver Painted Nylon Artificial Muscle Actuated by Joule Heating," *Proceedings of SPIE*, Vol. 9056, Paper No. 90560I, 2014.
16. Hiraoka, M., Nakamura, K., Arase, H., Asai, K., Kaneko, Y., et al., "Power-Efficient Low-Temperature Woven Coiled Fibre Actuator for Wearable Applications," *Scientific Reports*, Vol. 6, Article No. 36358, 2016.
17. Yip, M. C. and Niemeyer, G., "On the Control and Properties of Supercoiled Polymer Artificial Muscles," *IEEE Transactions on Robotics*, Vol. 33, No. 3, pp. 689-699, 2017.
18. Wu, L., de Andrade, M. J., Saharan, L. K., Rome, R. S., Baughman, R. H., and Tadesse, Y., "Compact and Low-Cost Humanoid Hand Powered by Nylon Artificial Muscles," *Bioinspiration & Biomimetics*, Vol. 12, No. 2, Paper No. 026004, 2017.



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