博 士 論 文 の 内 容 の 要 旨

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論文題目	Development of shape memory polyurethane actuator and its applications（形状記憶ポリウレタンアクチュエータの開発と応用に関する研究）

（博士論文の内容の要旨）

Shape memory polymers (SMPs) are a class of smart materials that can control shape by memorizing temporary shapes and returning to their original shapes. Particularly, shape memory polyurethane (SMPU) has become the favorite material for many desirable functions due to its high flexibility, high recovery, ease of processing, and low density. Especially, they are better in terms of biomaterials because they have biocompatibility and biodegradability. However, SMPU is an insulator material and has limitations for only thermal stimulus. To overcome this issue and interact with an electric response, an idea of electroactive polymer (EAP) was applied to conduct a dielectric elastomer actuator, which is a class of EAP used in many interesting applications, including artificial muscles, sensors. For integrating with biomaterials, Young's modulus of human skin or implants is generally less than 100 MPa, which is much lower than pure SMPU film's stiffness. Incorporating plasticizers such as dibutyl adipate (DBA) is an excellent candidate to enhance SMPU softness and perform gel-like soft films. Moreover, it can increase the polar attraction between the polymer and plasticizer, which contributes to improving the dielectric properties of SMPU composites. This dissertation develops SMPU gels focused on biomedical applications and demonstrates a new self-healing feature of soft actuators, which could activate both the thermal and electric stimulus. The main results are obtained as follows.

(1) The preparation of SMPU gels for soft actuators has two main factors to focus on, which are softness and dielectric properties. The softness of SMPU gels was improved close to a human implant, which is the energy dissipation factor of approximately 0.2–0.5, and the dielectric constant at 1 Hz in SMPU gels was significantly improved from 2.9 to 153 by the addition 2 times of DBA plasticizer, which allows the developed SMPU gels to function as sensor and actuators under applied electric fields with frequency dependence. The impedance control can allow for velocity control in robotic applications to imitate human motion. This could predict the further response of electric actuation.

(2) The actuation of electric and thermal was studied. Electric actuation of dielectric SMPU gel actuator was conducted with conductive materials charging, and its two-way shape deformation of contraction and expansion was investigated over several electric on-off cycles. The largest contraction reached 6.76% with an electric field of 3.42 V/μm. The thermomechanical analysis showed that SMPU gels could be stretched up to a high strain of 80% and showed immediately shape recovery behavior for the SMPU gels. The developed dielectric SMPU actuator displayed the ability to respond to thermal and electric stimuli. These composite materials have the potential for alternative actuation applications, including artificial muscle.

(3) Thermal actuation has been focused and adjusting the transition temperature of SMPU gels to activate the human body temperature range and design its applications for tube actuation. The plasticized SMPU tube was activated at 37 °C, faster than the pure SMPU tube. The effect of thermal triggering on tensile and viscoelastic properties was investigated, and according to thermomechanical analysis results, the shape recovery ratio of tensile deformation is up to 99%. Adding DBA plasticizers to SMPU demonstrated a quicker recovery rate than pure SMPU during tube compression and expansion on diameter direction. In addition, according to thermomechanical analysis results, the shape recovery ratio of tensile deformation is up to 99%.

(4) One of the most popular processing shapes for producing smart biomaterial devices is the tube actuator, especially for biomedical applications such as stents and artificial blood vessels, which can function in round shrinking and expansion deformation via diameter direction. We have fabrication two kinds of tube actuators. The first is developed for heat actuation. The tube compression and expansion demonstrated tube recovery in the diameter direction up to 83% for SMPU/DBA gel, and their large recovery behavior started from 37 °C. Their tube recovery acts with 30 s via 40 ºC. Another is developed for electric actuation. Cellulose nanocrystal (CNC) was incorporated into their composites to enhance the dielectric properties of SMPU and DBA composites to obtain a large deformation and fast response on electric actuation. The displacement on contraction and expansion was enhanced up to 11.58% on thickness direction. Furthermore, its tube contraction was occurred up to 800 µm (0.02% of its diameter direction).

(5) The biocompatibility in vitro experiment was conducted to address the possibility of biomedical applications. The cell adhesion showed that these gels have non-toxicity, and the cell proliferation demonstrated the cell growth on SMPU/DBA and SMPU/PEG. It indicated that they are not hindering cell growth. In addition, the cell adhesions and proliferation of NH3T3 mouse cells in vitro experiments supported their ability of biocompatibility.

As a total, we designed the developed SMPU, and the dielectric elastomer actuator displayed a multifunctional response to thermal and electric stimuli. For mechanical properties, the fast response, high recovery, and small stress were excellent features to be safe for use in the human body. These SMPU gel and its tube actuators can be used in the human body, which has aided our work on drug release systems for biomedical applications. In the future, SMPU gels are also expected to be used in producing thermal and electric tube actuators for microactuators, drug delivery, artificial blood vessels, and other aspects of tissue engineering.