博　士　論　文　の　内　容　の　要　旨

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学位授与年月日	2022 年 9 月 30 日
論文題目	Study on the functionality of thermoplastic epoxy and its applications (熱可塑性エポキシの機能性とその応用に関する研究)

（博士論文の内容の要旨）

In this study, the shape memory, thermal actuation, and self-healing functionality of thermoplastic epoxy resins are investigated, and their applications in artificial muscles, smart textiles, and flexible smart films are discussed. The main results obtained in this study are shown as follows.

(1) Development of thermoplastic epoxy filament for thermal actuation with excellent shape memory properties

An epoxy resin mixture composed of epoxy and phenol monomers is used to prepare thermoplastic epoxy polymer through a polymerization reaction and shape memory thermoplastic epoxy filament was successfully developed for the first time through a melt-drawing process. Tensile tests showed that the yield stress of the developed shape memory thermoplastic epoxy filament reached 63 MPa, which is an increase of 54% compared with thermoplastic epoxy films. Shape memory experiments showed that the developed shape memory thermoplastic epoxy filament has excellent shape memory performance, with a shape fixation rate of 97%, a shape recovery rate of over 97%, and good stability in cycling. Based on the shape memory performance analysis, the shape recovery stress of the shape memory thermoplastic epoxy filament was characterized. Shape recovery stress responded to temperature stability and increased with the increase of strain, reaching 1.45 MPa at a strain of 35%. The shape memory thermoplastic epoxy filament can reach an energy density of 0.066 J/cm3 during thermal actuation and shows greater application potential when processed into textiles. In addition, the chemical structure, thermal performance, and dynamic mechanical performance of shape memory thermoplastic epoxy filaments are analyzed. The developed shape memory thermoplastic epoxy filament shows excellent shape memory performance, and the output shape recovery stress is more than 5 times that of thermoplastic epoxy film, which provides huge application potential in the fields of artificial muscles and smart textiles.

(2) Development of shape memory thermoplastic epoxy films with human skin temperature response and self-healing function through the regulation of thermal and mechanical properties by PEG

In order to expand the application scope of thermoplastic epoxy resins, epoxy-polyethylene glycol (PEG) films with controllable thermal and mechanical properties were prepared by dispersing PEG, and their multifunctional shape memory properties were explored. PEG has excellent compatibility with unpolymerized epoxy resin because of the existence of a large number of C-O bonds and hydroxyl groups. A uniform and stable epoxy-PEG colloidal-like mixture can be obtained through environmentally friendly melt dispersion, and the PEG molecules are uniformly and stably wrapped in the polymerized epoxy molecular chain. The dispersion of PEG with different contents enables epoxy-PEG films to be regulated between rigid and flexible without significantly reducing the thermal stability of epoxy-PEG, which greatly expands the application range of epoxy resin. Epoxy-PEG films with different PEG contents all have excellent shape memory properties with shape recovery rates between 92% to 100%, and can be regulated by heat-stimuli between original and temporary shapes. In addition, based on the regulation of Tg of epoxy-PEG films by PEG, the heat-stimuli temperature of epoxy-PEG films can also be adjusted to meet different application requirements. With increasing PEG content, epoxy-PEG films are able to achieve shape memory with human skin temperature response, self-healing, and heating-based adhesive functions. The epoxy-PEG films with controllable thermal and mechanical properties prepared by dispersion of PEG show greater application potential in the fields of shape memory and flexible smart materials.

(3) Development of shape memory thermoplastic epoxy filament with controllable heat-stimuli temperature for adapting thermal actuation of different temperature responses

Shape memory thermoplastic epoxy-polyethylene glycol (PEG) filaments were prepared for the first time based on the research of PEG-modified thermoplastic epoxy, and the heat-stimuli controllability of shape memory function was innovatively endowed by adding PEG in an environmentally friendly way. PEG regulates the Tg of PEG-modified thermoplastic epoxy without interfering with the polymerization reaction of epoxy. The melt spinning temperature of PEG-modified thermoplastic epoxy is also reduced from 300 °C to 220 °C, and the spinning effect is improved. After PEG modification, Young's modulus of PEG-modified thermoplastic epoxy filaments drawn from PEG-modified thermoplastic epoxy pellets was higher at room temperature. The developed PEG-modified thermoplastic epoxy filaments have excellent shape memory effect with high shape recovery rates of 93.1%–99.0%, and shape fixation rates ranging from 95.8% to 99.1% are also improved after PEG modification. Furthermore, PEG-modified thermoplastic epoxy filaments have controllable stimuli temperatures of shape memory ranging from 69 °C to 105 °C. Additionally, the recovery stress of PEG-modified thermoplastic epoxy filaments after PEG modification was increased to 1.77 MPa, and the recovery temperature of the maximum recovery stress is reduced from 97 °C to 58 °C as PEG content increases to 15 wt%. The problems of difficult melt processing and uncontrollable stimuli temperature for thermoplastic epoxy were solved in this study. The developed PEG-modified thermoplastic epoxy filaments with heat-stimuli controllability of the shape memory function demonstrated excellent thermal actuation capabilities and have shown greater application potential in thermal actuation, smart textiles, and artificial muscles.

In conclusion, from the development of thermoplastic epoxy shape memory filaments to the development of multifunctional epoxy-PEG including shape memory by dispersing PEG, and further to the development of thermally controlled shape memory epoxy-PEG, this research developed multifunctional thermoplastic epoxy-PEG material in an environment-friendly and low-cost way base on the revealing the dispersing mechanism of PEG in thermoplastic epoxy. In terms of textiles, the developed pure thermoplastic epoxy filaments show great thermal actuation ability on the basis of excellent shape memory properties. The further developed thermoplastic epoxy-PEG filaments can respond to heat-stimuli at different temperatures on the basis of maintaining the excellent shape memory properties of pure epoxy filaments, showing an excellent controllable heat-stimuli shape memory function. In terms of film materials, the developed thermoplastic epoxy-PEG films with more PEG content showed a change from rigid to flexible with increasing PEG content and showed shape memory with human skin temperature response, self-healing, and adhesion performance, which was extremely greatly improved the application range of thermoplastic epoxy.