STRETCHABLE EUTECTOFIBERS VIA ROTARY WET-SPINNING FOR WEARABLE STRAIN SENSORS

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ABSTRACT

Eutectofibers represent a significant advancement in flexible electronics, addressing the limitations of current conductive fibers such as function loss and deformation by drying. By directly spinning an alginate/polyethylene glycol diacrylate (PEGDA) /Irgacure 2959 aqueous solution into a calcium chloride ($CaCl_2$) dissolved in a natural deep eutectic solvent (NADES) bath, we can achieve high throughput fabrication in a fast manner. The resulting eutectofibers exhibit exceptional flexibility and transparency, surpassing the performance of traditional conductive fibers. Furthermore, the eutectofibers exhibit superior anti-freezing property and mechanical performance. In this study, we introduce a large-scale production of eutectofibers with rotary wet spinning system, offering a promising pathway for the development of next-generation flexible electronic devices. The scalability of this fabrication method opens doors to the widespread adoption and implementation of eutectofibers in diverse applications. One promising application of eutectofibers as a strain sensor is exemplified.

KEYWORDS

Rotary wet-spinning, Eutectofiber, Wearable Strain sensor

INTRODUCTION

Stretchable conductive fibers play a crucial role in advancing the field of next-generation flexible electronics[1]. Current electron-based conductive fibers are often opaque and relatively rigid and experience a significant increase in resistance when stretched. To overcome these limitations, there has been a notable shift towards soft, stretchable, and transparent ion-conductive hydrogel fibers [2, 3]. However, the high water absorbency of hydrogels makes them highly sensitive to environmental conditions, particularly humidity and temperature. By the air exposure, the mechanical properties such as stretchability and elasticity of hydrogel can be changed leading to a loss of ability as a strain sensor [4]. These challenges are also creating another challenge in the manufacturing process, the sensitive properties to various environmental factors of hydrogel make it hard to achieve uniformity and reproducibility in different fabrication batches. Therefore, challenges in manufacturing and susceptibility to drying and freezing have hindered the widespread application of hydrogel fibers [2, 5]. In response to these challenges, eutectogel fibers (eutectofibers) have been developed to address the drawbacks of traditional hydrogel counterparts. Herein we introduce a eutectogel-based strain sensor. Eutectogel maintains the softness of hydrogel while exhibiting higher stability under external environmental conditions. The fabrication method based on the wet spinning system not only enables high throughput fabrication but also ensures consistent fiber properties, such as diameter and porosity, crucial for their performance as stretchable conductive materials [6]. Moreover, the continuous rotary wet spinning method allows for simultaneous injection and curing of the fibers, leading to rapid fabrication. This innovative approach not only overcomes the challenges of drying and freezing susceptibility but also paves the way for the scalable production of eutectogel fibers for a wide range of applications. Given the high ionic conductivity of the eutectofibers, one can utilize the fibers as conductive threads for flexible wearable electronic applications. Their sensitivity to strain also makes them ideal as strain or pressure sensors that can be used to monitor physical activity. Preliminary results indicate promising performance as a strain sensor.

DISCUSSION

Eutectofiber preparation

Our innovative one-pot fabrication approach involves the continuous rotary wet-spinning of eutectofibers, as depicted in figures 1(A) and 1(B). The rotary wet spinning method is based on the formation of fibers through a physical and covalent hybrid crosslinking mechanism (Figure 1C), followed by a solvent exchange process. Specifically, we directly spun an alginate/polyethylene glycol diacrylate (PEGDA)/Irgacure 2959 aqueous solution into a bath containing calcium chloride ($CaCl_2$) dissolved in a natural deep eutectic solvent (NADES) comprising choline chloride (ChCl) and ethylene glycol (EG) at a molar ratio of 1:2.5. Simultaneously, UV light ($\lambda = 365$ nm) is exposed to the fibers to promote the covalent photocrosslinking of PEGDA polymeric chains. Subsequently, the fibers undergo solvent exchange by keeping them immersed in the $CaCl_2$/NADES bath. By injecting the alginate/polyethylene glycol diacrylate (PEGDA)/Irgacure 2959 aqueous solution through the syringe on one side and curing with UV light simultaneously, we can achieve fast fabrication.

Hydrogel fiber and eutectogel fiber comparison

To optimize the composition and compare the propertied of 4 different compositions including alginate-based hydrogel, and eutectogel with and without PEGDA were prepared. The alginate eutectofibers were prepared using the same protocol, whereas comparative hydrogel fibers were prepared using $CaCl_2$ aqueous solution instead of $CaCl_2$/NADES. As shown in Figure 2(A), gravimetric analysis revealed that adding PEGDA to the fibers resulted in superior weight stability, with the fibers retaining their weight for 48 hours, whereas fibers without PEGDA lost approximately 75% of their weight within 4 hours. This highlights the importance of PEGDA in enhancing the stability of eutectogel fibers over time. To evaluate the electrical conductivity of fibers, impedance measurements were conducted in the range of 0 to 8 MHz using an LCR meter (IM3536, Hioki, Japan). As shown in Figure 2(B), hydrogel fibers exhibited constant impedance for the overall frequency range. In contrast, eutectofiber impedance magnitude decreased with increasing frequency, indicating its suitability for high-frequency signals. This behavior is advantageous for applications requiring responsive and adaptable conductive materials. Based on these results, alginate/PEGDA eutectogel fiber proved superior weight stability and frequency-dependent impedance characteristics making it a promising candidate for use in next-generation flexible electronics and strain sensing applications.

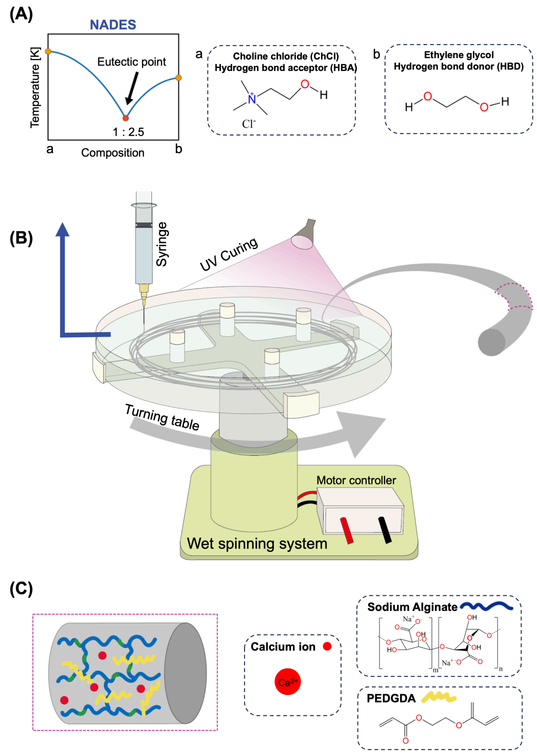

Figure 1 Schematic representation of the stages of eutectofiber fabrication via the rotary wet-spinning process: (A) synthesis of natural deep eutectic solvent (NADES) using choline chloride and ethylene glycol; (B) spinning of aqueous alginate/PEGDA solution over $CaCl_2$/NADES solution; (C) hybrid (ionic and covalent) crosslinking of fiber.

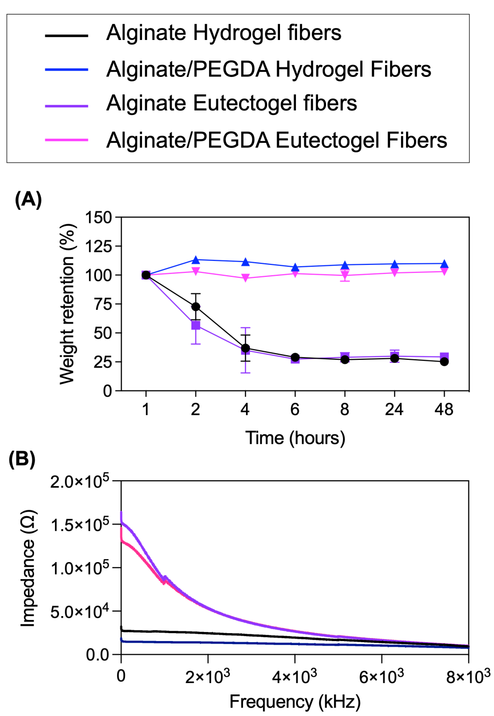

Figure 2 (A) Weight change of eutectofibers and hydrogel fibers at 20 °C and 33% humidity. (B) Impedance measurement of the fibers.

Optical property of eutectofiber

The photographs presented in Figure 3 showcase the remarkable transparency and flexibility of alginate/PEGDA eutectofibers. In Figure 3(A), an optical microscope (VHX 7000, Keyence, Japan) captures the alginate/PEGDA eutectofiber placed on the fingers of a glove. The thin, single eutectofiber exhibits bending in accordance with the finger's movement, highlighting its exceptional flexibility and conformability to dynamic shape. Furthermore, the high stretchability and conformability of alginate/PEGDA eutectofiber were assessed under extreme deformation. As shown in Figure 3(B), the eutectofiber is tied into an overhand knot, showcasing its excellent knitting feasibility and conformability without any evident fractures or damage.

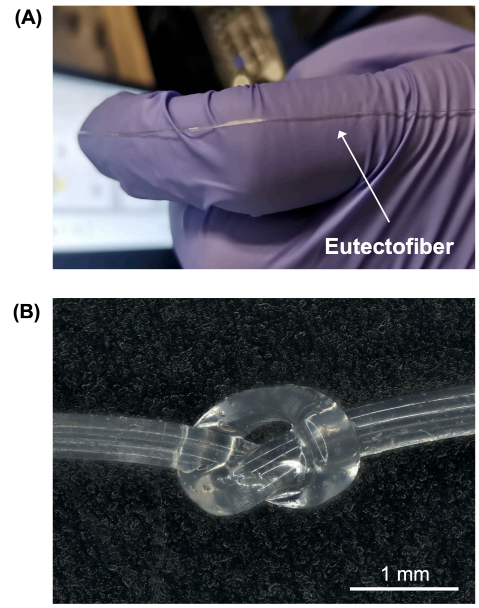

Figure 3 (A) Photographs of a long single collected alginate/PEGDA eutectofiber and (B) eutectofiber knot.

Antifreezing property of eutectofiber

Notably, eutectofibers exhibited exceptional anti-freezing properties and maintained stable mechanical performance even at subzero temperatures, maintaining high transparency and stretchability. To confirm the anti-freezing property of alginate/PEGDA eutectofibers, their stretchability was compared between samples restored at room temperature and those restored at -80°C. As depicted in Figure 4A, the sample restored at room temperature shows a stable stretching property. Similarly, the sample restored at -80°C also exhibited comparable stretchability, as depicted in Figure 4(B). This observation highlights the remarkable anti-freezing characteristics of alginate/PEGDA eutectofibers.

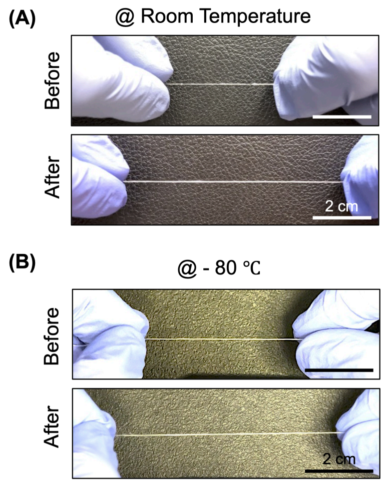

Figure 4 Antifreezing property and stability of alginate/PEGDA eutectofibers, demonstrating that the fiber retained high stretchability at −80 °C.

Electrical characterization of eutectofiber

To assess the sensitivity and mechanical durability of alginate/PEGDA eutectofibers, impedance changes were measured during repeated stretching and releasing deformations. As depicted in Figure 5, 20% strain for 1 second and the releasing cycle was repeated four times, and impedance was measured at 8 MHZ. During each cycle of repeated 20% stretching, the relative impedance showed an increase ranging from approximately 1.22 to 1.30 times higher than the initial impedance in all four stretching cycles. This behavior demonstrates the sensitivity of eutectofibers to mechanical deformation and their ability to detect rapid cyclic motions effectively as promising candidates for the development of strain sensors for monitoring physical activity similar to our prior work [7-9].

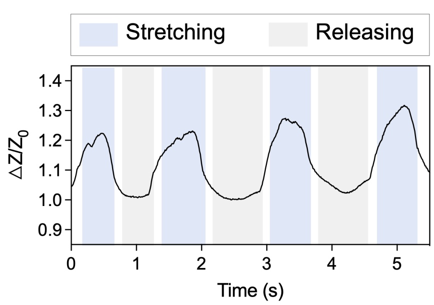

Figure 5 Relative impedance change measurement under 20% stretching and releasing cyclic deformation.

CONCLUSION AND FURTHER WORK

We have successfully demonstrated the fabrication of eutectogel fibers using the continuous rotary wet spinning system. This method allows for fast and mass-production of fibers by directly injecting the alginate/PEGDA solution into a bath containing CaCl2 dissolved in NADES. The fabricated alginate/PEGDA eutectofiber demonstrated superior weight stability, transparency, and flexibility. Furthermore, our study confirmed the remarkable anti-freezing properties of alginate/PEGDA eutectofibers through stretchability testing at subzero temperatures. This highlights their potential for reliable performance even in cold environments. Additionally, impedance measurements during repeated stretching and releasing deformations validated their sensitivity to mechanical deformation, indicating their suitability for dynamic applications. As a further study, we propose exploring the integration of eutectofibers into electronic devices and systems allowing them to demonstrate real-world applicability, particularly in areas such as movement monitoring where the sensitivity to mechanical changes could be leveraged effectively.

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