Title	多孔質構造の導入によるポリベンズイミダゾールフィルムの性能向上
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Abstract

Polybenzimidazole (PBI) is a high-performance polymer known for its excellent thermal stability, mechanical properties, and chemical resistance, making it a material candidate in demanding environments. In particular, poly(2,5-benzimidazole) (ABPBI) is the simplest chemical structure of PBI, consisting of repeated benzene and imidazole rings. However, several challenges of ABPBI remain such as low sustainability and high-cost availability caused by the commercial time-consuming polycondensation process, heterogeneity and low flexibility of processed film due to the rigid backbones of ABPBI, lack of processability with insolubility in common solvents, and moisture sensitivity which limited their further application as a super engineering plastic.

One of the primary challenges facing the widespread adoption of ABPBI films is the high cost of production. The complex synthesis and processing techniques required to produce ABPBI contribute to its higher price point compared to other polymers. Moreover, scaling up the production of ABPBI films while maintaining their unique properties with crucial uniformity is also required in manufacturing technologies. In Chapter 2, ABPBI was synthesized successfully by using Eaton`s reagent, which is a time-saving polycondensation procedure. Notably, the adopted monomer is bio-derivable, which remarkably improves sustainability and reduces the production cost of ABPBI. The fabricated ABPBI film by solution casting method showed heterogeneous and featured branched patterns of thick brown regions, which may have originated from the surface condensation of ABPBI to form sticky fibrous aggregates. Subsequently, a hard-templating method using silica nanoparticle fillers was employed to homogenize the ABPBI film. The composite ABPBI films had reduced heterogeneity and roughness compared with the original ABPBI film. Herein, a formation mechanism could be attributed to the ABPBI aggregate stuck to the surfaces of the silica nanoparticles, which were well dispersed over the film by ultrasonication. The reduced cost of production and improved uniformity suggest the potential of ABPBI as a material for a broader range of industry uses, e.g., fuel cell separators, leakproof films, and filtration membranes.

Although ABPBI is strong and rigid, it can be relatively brittle as well. Its low elongation at break limits its use in applications where flexibility or toughness is required. In Chapter 3, another bio-derivable monomer was incorporated into the ABPBI backbones named poly(BI-*co*-A) to develop flexibility. Poly(BI-*co*-A) was synthesized using viscous poly(phosphoric acid). Moreover, to further enhance the durability, flexibility, and performance of poly(BI-*co*-A) film, porous structures were constructed via a silica-etching method. The prepared porous poly(BI-*co*-A) films showed much higher toughness and mechanical stability, which could be expected to play a crucial role in enabling the next generation of durable and resilient electronic devices, including their potential use in high-performance sensors, smart textiles, and flexible batteries. On the other hand, poly(BI-*co*-A) film has some degree of water absorption, which can affect its mechanical properties over time, especially under humid conditions. This could be problematic in applications where dimensional stability or mechanical strength must be maintained in moist environments. Consequently, dehydration treatment of silica nanocomposite poly(BI-*co*-A) films in an electric furnace was employed to improve the surface wettability of the poly(BI-*co*-A) film by decreasing the hydroxy bonds in the films. The fabricated silica nanocomposite poly(BI-*co*-A) films displayed an enhanced thermal resistance, even comparable to some metals, implying their replacement of heavyweight metal or inorganic materials. Nevertheless, the improved wettability of poly(BI-*co*-A) films further ensures their potential candidate for applications in an extreme environment such as fuel cell membranes, barriers, and anti-fouling film, aircraft, and spacecraft.

Regarding the future scope of this work, the successful PBI films with enhanced performance such as low-cost procedure, sustainable resources, high uniformity, improved flexibility, and enhanced toughness, have significant potential in a variety of industries, from energy and aerospace to electronics and environmental protection. Whether through their role in improving fuel cell performance, protecting advanced electronic devices, or enabling cleaner industrial processes, PBI films are set to play a key role in the future of materials science and engineering. The ongoing advancements in manufacturing, sustainability, and application development will further expand the use of PBI films, making them critical materials for the 21st century and beyond.

Key words: Heat-resistant polymers, polybenzimidazole, porous structures, homogenization, high toughness