【Grant-in-Aid for Specially Promoted Research】

Solid-State Materials Science of Supramolecular Polymers and Their Applications

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Project Information	Project Number : 23H05408 Project Period (FY) : 2023-2027 Keywords : Solid State Supramolecular Polymers, Solid State Properties, Phase Separation, Entropy, Dynamics

Purpose and Background of the Research

●Outline of the research

Waste plastics have led to significant environmental concerns. However, society remains reliant on inexpensive and convenient plastics. Supramolecular polymers offer a promising solution to this problem. This research project aims to develop "innovative polymer materials for the SDGs era" that possess exceptional solid-state properties not achievable with traditional covalent polymers. We will investigate the "solid-state materials science of supramolecular polymers" to create novel functional materials that possess unparalleled mechanical strength and can be easily converted back into raw materials under specific conditions. We will elucidate the correlation between structure, dynamics, and properties of supramolecular polymers in the solid phase, connecting the missing link between conventional polymer science and the emerging field of supramolecular polymer science (Fig. 1).

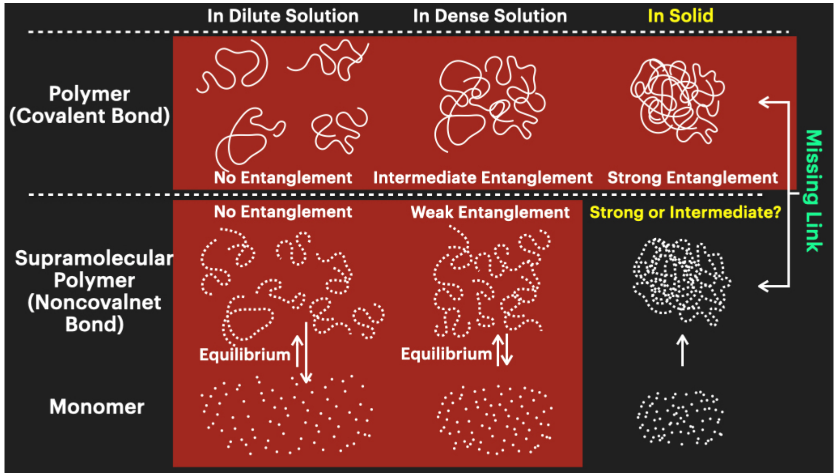

Figure 1. Schematic diagram of the missing link between conventional polymer science and supramolecular polymers and Structure-Dynamics-Property correlations.

●Originality of the research

We discovered that some short-chain polymers can achieve the incompatible qualities of "mechanical robustness" and "self-healing ability at room temperature" by utilizing specific hydrogen bonds. Furthermore, we have revealed that some hydrogen-bonded supramolecular polymers possess a substantial Young's modulus comparable to acrylic resins. The misconception that "supramolecular polymers are inherently dynamic and weak" stems from their behavior in solution, but it is not a universal characteristic. By controlling the processes of bond formation and dissociation, even in non-covalently bonded monomers, we can develop supramolecular polymers with extraordinary mechanical properties.

Expected Research Achievements

●Research plans

To investigate the relationship between structure, dynamics, and properties of solid-state supramolecular polymers, this study accomplish three primary objectives.

(1) Development of robust yet recyclable supramolecular polymer glasses (SPGs): We will explore supramolecular polymer materials that exhibit superior robustness compared to engineering plastics. This will be achieved by simply blending multiple small organic molecules capable of forming salt bridges with each other. They will undergo phase separation in water, and subsequent drying will yield transparent SPGs with an exceptional Young's modulus. By expanding the range of small organic molecules, we will diversify the basic structure and physical properties of SPGs.

(2) Development of room temperature self-healing SPGs: We have developed self-healing polymer glasses (Fig. 2). By utilizing small organic molecules containing multiple thiourea groups that form dense hydrogen bonds without inducing crystallization, we will develop a series of supramolecular polymers with high strength and room-temperature self-healing properties. This approach enables the creation of supramolecular polymers with diverse structures and physical properties.

(3) Phase separation of precisely synthesized supramolecular block copolymers: The phase separation behavior of supramolecular block copolymers remains largely unexplored due to the challenges associated with synthesizing supramolecular polymers with precisely controlled chain lengths. By supramolecular polymerization of two types of hydrogen-bonded monomers with hydrophilic and hydrophobic side chains, we synthesized supramolecular block copolymers with different structures and lengths. Through electron microscopy and relaxation time measurements using solid-state NMR, we will investigate the phase separation behavior and mechanical properties of these supramolecular block copolymers in the solid phase (Fig. 3).

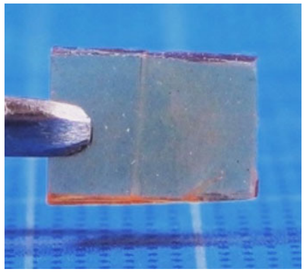

Figure 2. Photograph of self-healing polymer glass.

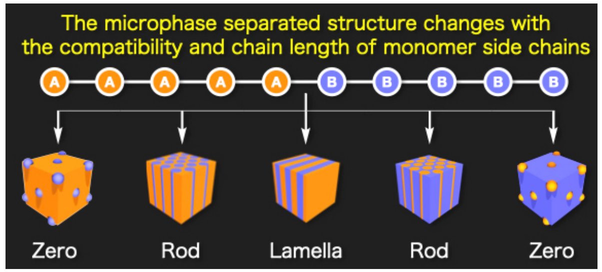

Figure 3. Schematic diagram of microphase separated structures of supramolecular block copolymers.

●Expected research achievements and scientific significance

The novel research direction pursued by this project is anticipated to attract numerous talented researchers who were previously hesitant to enter this field. As a result, the field of "solid-state materials science of supramolecular polymers" is poised for significant growth and development. The collective efforts of this project and newly engaged researchers will shed light on the potential of supramolecular polymers as solid materials suitable for practical applications. By striving to replace 25% of all polymer materials with renewable natural alternatives and an additional 25% with materials derived from supramolecular polymers, a substantial improvement in the global plastic waste predicament can be achieved. This paradigm shift will mark an essential milestone towards a sustainable future for humanity.

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