Ultrahighly Refractive and Degradable Thiourea Polymers with Dense and Multiple Intermolecular Hydrogen Bonds

(*Dept. of Applied Chem., Waseda Univ.*) ○Seigo Watanabe, Kenichi Oyaizu
Keywords: Thiourea; High Refractive Index; Hydrogen Bonds: Degradability

High-refractive-index polymers (HRIPs) are attractive for preparing high-performance and flexible optical devices, and the required value of the refractive index (RI) has been getting higher up to over 1.8.[1] Although there has been much effort on enhancing the RI of polymers such as sulfur-rich molecular design[2] and introducing heavy chalcogenide atoms,[3] few HRIPs satisfying both RI over 1.8 and visible transparency have been reported. As a solution, we recently reported hydrogen-bond-incorporated poly(phenylene sulfide)s representing ultrahigh RI (n_D = 1.85 in maximum) which was ascribed to the free volume reduction induced by intermolecular H-bonds.[4]

In this study, we report ultrahighly refractive aromatic thiourea polymers as an expanded version of our strategy (Figure 1a). High-molecular-weight thiourea polymers were easily prepared via polyaddition or polycondensation (Figure 1b). As thiourea form randomly distributed hydrogen bonds,[5] the polymer films were flexible and transparent despite the presence of dense hydrogen bonding networks (Figure 1c). The polymers showed high refractive indices of n_D ~ 1.7-1.8 owing to the dual contribution of high polarizability and dense multiple hydrogen bonds. We also demonstrated that these polymers were degradable under the presence of excess diamine owing to the dynamic properties of thiourea units.

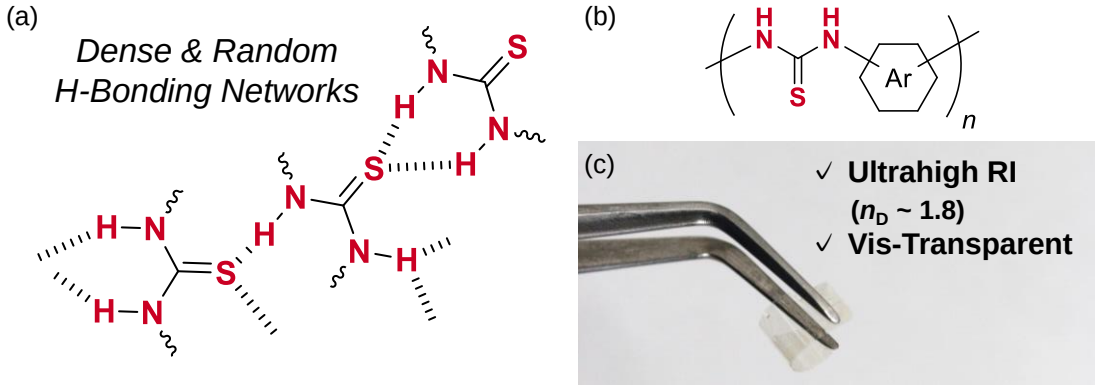

Figure 1. Features of thiourea polymers in this study: (a) Schematic representation of hydrogen bonding networks by thiourea. (b) Molecular design of the thiourea polymers. (c) Photograph of flexible thiourea film with ultrahigh RI and visible transparency.

1) T. Higashihara, M. Ueda, *Macromolecules* **2015**, *48*, 1915. 2) J. Pyun *et al.*, *ACS Macro Lett.* **2020**, *9*, 245. 3) J. Zhu *et al.*, *Polym. Chem.* **2019**, *10*, 4279. 4) S. Watanabe, K. Oyaizu *et al.*, *Macromolecules* **2022**, *55*, 2252.; *ACS Polym. Au* **2022**, *2*, 458. 5) T. Aida *et al.*, *Science* **2018**, *359*, 72.

© The Chemical Society of Japan