

A tough polyurethane elastomer with self-healing ability

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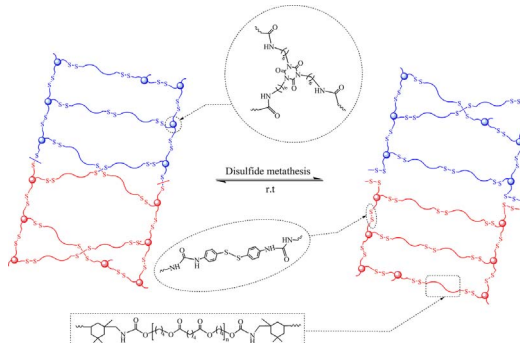
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ABSTRACT

Due to sufficient polymer chain mobility and molecular diffusion, most of the present intrinsic self-healing polymers that heal damage by utilizing reversible bonds are soft or gel. However, a tough self-healing polymer is needed as they can be used as strong and light weight material for engineering applications. Based on the high versatility of polyurethanes, herein we report the development of a disulfide-containing poly(urea-urethane) (PUU) network, where aromatic disulfide diamine is used to establish linkages for tri-functional homopolymer of hexamethylene diisocyanates (tri-HDI) as well as aliphatic prepolymer chains, resulting in the readily conversion between a well cross-linked network and an un-crosslinked structure. With this linkage, the network is able to balance the two seemingly contradictory forces, exhibiting simultaneously superior mechanical performances and high self-healing efficiencies in the absence of any catalyst or external intervention. In addition, these materials are capable of repairing themselves repeatedly thus possessing a high potential for industrial applications with further investigation.

GRAPHICAL ABSTRACT



1. Introduction

Self-healing polymeric materials are regarded as an emerging class of smart materials, due to their ability to partially or fully repair the internal damage inflicted on them, thereby extending the lifetime of the material in numerous applications without the need for detection or manual intervention of any kind [1–2]. The initial method to prepare a polymeric material is based on encapsulation concept proposed by White et al. [3]. They embedded microcapsules containing reactive

healing agent into the polymer matrix. When a crack propagates through the material, it causes the release of the healing agent from the capsule into the crack plane where it solidifies and repairs the material. The capsule-based approach has since been widely studied in a broad range of disciplines from bio-medical materials to protective coatings [4–6]. The disadvantages of such extrinsic self-healing method that operates with the aid of compartment are quite complex and unavailable for repeatable healing response. Inspired by blood vessels in biology, an alternative approach to achieve self-repair polymeric

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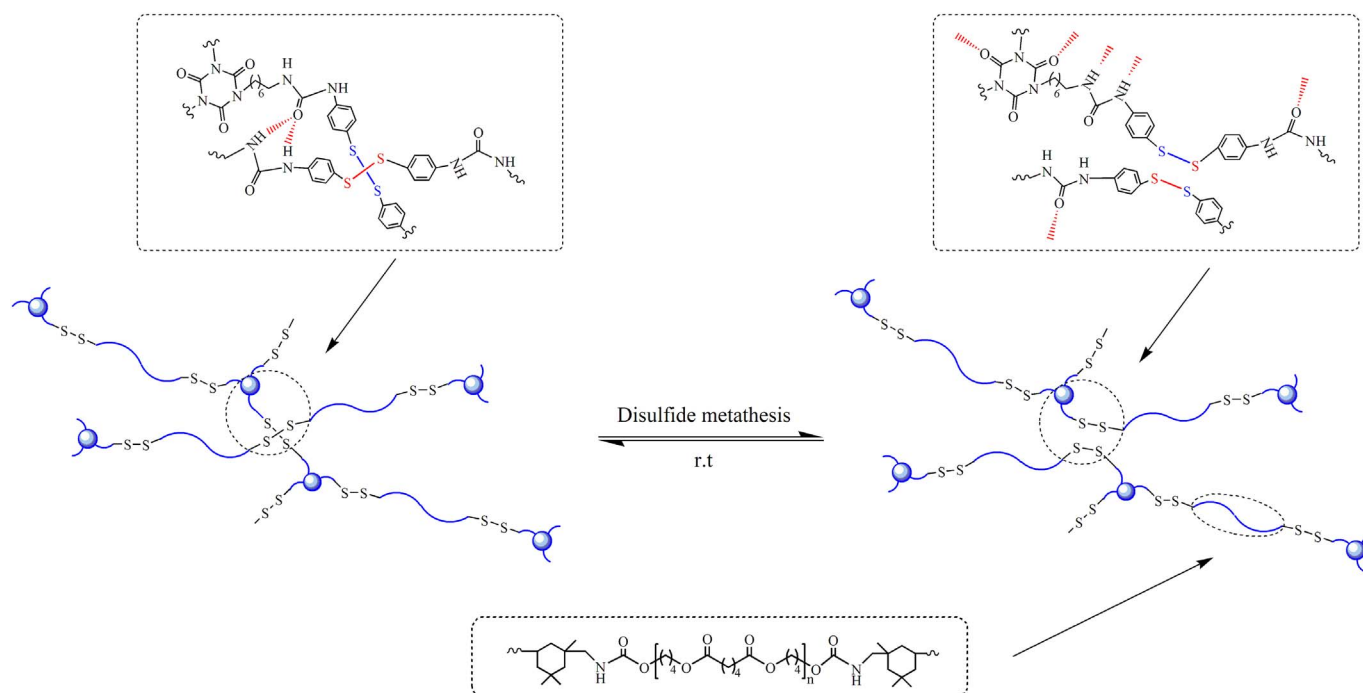


Fig. 1. Healing mechanism of poly(urea-urethane) with disulfide metathesis.

materials is the incorporation of microchannels within the polymer. With repeated self-healing abilities, however, vascular self-healing materials, along with the capsule-based ones, usually have compromised initial properties which are primarily affected by the embedding. A better method to extend the lifetime of a material is to utilize the dynamic reversible covalent bonds [7–9] or macro-molecular interactions (noncovalent bonds) [10–12] contained in the materials, which is classified as intrinsic self-healing. The research and potential application of this material is currently becoming the subject of increasing interest due to their ease of applicability and their potential for mass production [2,13,14].

Wool et al. dedicated to the mechanism study of thermoplastic intrinsic self-healing polymers and pointed out that the healing process goes through several phases: (i) surface rearrangement, (ii) surface approach, (iii) wetting, (iv) diffusion and (v) randomization [15,16], during which the longitudinal chain diffusion is responsible for crack healing [17]. There is no denying that polymer chain mobility is a key factor to facilitate self-healing. In addition, it is worth noting that whereas craze healing occurs at temperatures above and below the glass transition temperature (T_g), crack healing happens only at or above the T_g [18]. The chain mobility required for intrinsic self-healing is achieved either by heating the polymeric material above its T_g , or using solvents to promote depression of the effective T_g to below room temperature [19]. When it comes to self-healing under mild conditions where no stimulus but a slight heat was applied to the system, low T_g of the free segment itself is the basic premise of sufficient chain mobility attributed to high self-healing efficiency, which means the material has to be very soft or has relatively low-molecular weight. This explains the reason why intrinsic self-healing materials that can be healed under

mild conditions always have compromised mechanical properties [9,12,20,21]. It is a challenge to achieve self-healing in tough polymers without the intervention of any kind owing to its reduced molecular segment mobility. As successful as these intrinsic healing techniques were in laboratory, the scope of self-healing polymers is limited when it comes to engineering applications due to their poor mechanical performances. Therefore it is imperative to design self-healing polymers with high healing efficiency, strong mechanical properties and light weight, for applications in fields such as aviation and construction industry [22].

The intrinsic self-healing polymers enable crack healing mostly under certain stimulation, such as pH [20,21] or a source of heat [23,24] or light [25,26], determined by specific molecular structures or reversible reactions applied to the polymers. The metathesis reaction of aromatic disulfides is known to transfer readily at room temperature without the need for any stimulation like catalyst or light [27–29], thus providing a mechanism for self-healing under mild conditions. Combining aromatic disulfide metathesis and the great versatility of polyurethane, disulfide bonds were incorporated into PUU networks by reacting 4,4'-dithiodianiline (DTDA) with tri-HDI, along with aliphatic prepolymer chains synthesized from isophorone diisocyanate (IPDI) and poly(butyladipate) diol (PBA). With this method, disulfide bonds were introduced and well-distributed surrounding the cross-linking points of tri-HDI by structural tuning (Fig. 1). On the one hand, a well cross-linked network has been constructed as we can see on the left. The six-membered ring of tri-HDI, a rather stable structure which can provide the PUU network with a tough skeleton, is the key to bringing the material's superior mechanical properties as the cross-linking center. On top of that, the quadruple hydrogen bonds, formed between

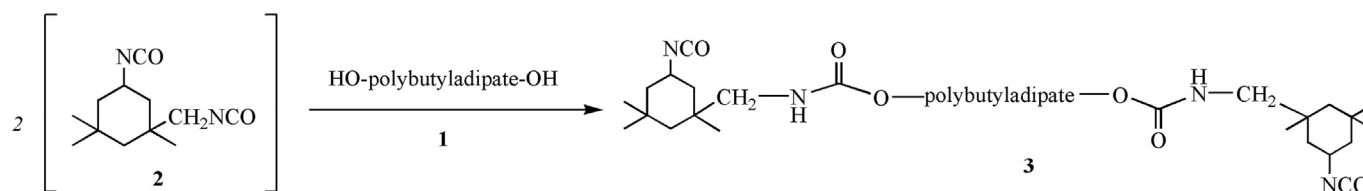


Fig. 2. Synthesis of the isocyanate-terminated prepolymer 3.

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