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Review

Biodegradable polyester shape memory polymers: Recent advances in design, material properties and applications

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ABSTRACT

Shape memory polymers (SMPs) is a class of well-studied smart materials with a variety of applications. In recent years, biodegradable polyester SMPs have received increasing attention from the research community both as academic interest and for their potential usefulness in biomedicine and soft robotics. In this contribution, the most recent progress in the area of biodegradable polyester-based SMPs is summarized with regards to their structural designs, thermomechanical properties, shape memory performance and some emerging characteristics, followed by a discussion on the specific applications of these SMPs in biomedicine and soft robotics. The prospective research direction for the future development of polyester SMPs as advanced functional materials is also discussed.

1. Introduction

Shape memory polymers (SMPs) have received an increasing attention in recent years due to their ability to change from one conformation (temporary shape) to another (permanent shape) when exposed to a specific environmental trigger (temperature, light, etc.) [1-7]. The physical and chemical mechanisms by which these shapememory behaviours are triggered include melting/glass transitions, reversible covalent/supramolecular interactions, photo-induced bondings, change in hydrophilicity, bondings facilitated by specific ions, etc. [8–11]. Such materials have tremendous potential in applications in which the actuating material has to be flexible and shape-conforming, while still possess sufficient mechanical strength. Common examples of such applications include the shape-adaptive industrial grips and autochoke element for engines [12,13]. While a number of polymeric shape-memory materials have been developed and work well for their intended applications, they also possess varying degrees of biotoxicity as well as resistance to biodegradation, thus severely limiting their usefulness for biomedical applications. An example is the Ni-Ti shapememory surgical alloys [14-16] which are biocompatible but does not degrade in-vivo due to their metallic nature, thus requiring invasive removal procedures once their usefulness expires. Therefore, many ongoing research efforts in polymer and material chemistry aim at incorporating shape-memory characteristics into biocompatible and environmentally-friendly polymers, and there are many reviews summarizing endeavours in this area [17-19].

Polyesters are among the earliest and most extensively investigated

biodegradable soft materials [20-22]. For instance, poly(lactic acid) (PLA), poly(glycolic acid) (PGA), poly(caprolactone) (PCL) and a range of their copolymers have been extensively researched as synthetic biomaterials in sutures, plates and fixtures for fracture fixation devices, scaffold for cell transplantation and other clinical applications [23–28]. Polyesters are easily available from biomass sources for mass production. Polyesters can be developed synthetically from polycondensation of various combinations of diols and dicarboxylic acids, self-polycondensation of hydroxyl acids and ring-opening polymerizations (ROP) of lactones and lactides [29-32]. ROP, in particular, is very popular in polyester synthesis since it is efficient, generates no side products (such as water in the case of polycondensation) and requires relatively mild reaction conditions [33]. Some of the most extensively studied biodegradable polyesters include PGA, PLA, their copolymer poly(lactide-co-glycolide) (PLGA), PCL, Poly(ω-pentadecalactone) (PPDL), poly(hydroxyalkanoates) (PHA) such as poly(3-hydroxybutyrate) (PHB), poly(dioxanone) (PDO, PDX or PDXO) and poly (ethylene succinate) (PES) [34-37]. These biodegradable polyesters have found extensive applications in delivery carriers in diversified formations, target specific delivery, as well as scaffold based tissue engineering and mimicking natural extracellular matrix (ECM) [38-41].

The greatest advantage of polyesters is their excellent biocompatibility and controllable degradation by simple hydrolysis in aqueous environments such as body fluids [42–48]. Polyesters, particularly the aliphatic polyesters, are one of the only high molecular weight compounds that have been proven to be hydrolysable due to their extremely

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hydrolysable backbone [49–53]. These backbones can fit into the active sites of hydrolases which speed up the degradation of these polymers [54–57]. Furthermore, the degradation products will ultimately be metabolized to carbon dioxide and water or excreted through the kidney [58,59], therefore presenting very little toxicity. In the light of these characteristics, PGA, PLA and many other common polyesters have already been approved for medical use by regulatory bodies such as the US Food and Drug Administration (FDA) [60–62].

With their excellent biocompatibility, well-defined glass-transition temperatures and functional versatility, polyesters appear to be very promising backbone material when designing shape-memory polymer systems, especially for the "soft" segments in thermal transition-based SMPs [63,64]. This review first focuses on the most recent advances in polyester-based SMPs in terms of material/structural designs, thermal/ mechanical properties, shape-memory performance and biocompatibility, followed by the discussion on the latest advances in applications of polyester-based SMPs, particularly in 1) biomedical applications where SMPs with excellent biocompatibility and shape-memory transitions temperatures near body temperature are most useful, and 2) soft actuators where the mechanical strength, shape recovery consistency and shape memory reversibility are more important. The potential usefulness of polyester SMPs for other applications such as smart switches will also be discussed (Scheme 1). Table 1 gives a brief summary of the properties and applications of polyester SMPs to be discussed in this review.

2. Biodegradable polyester SMPs

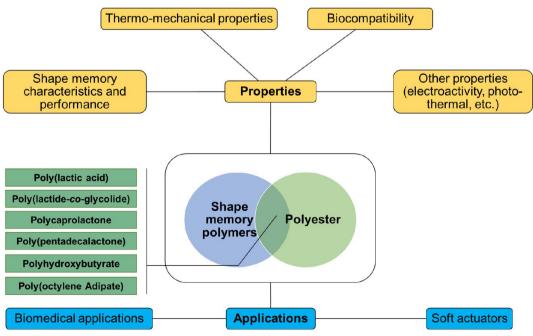
2.1. Poly(lactic acid) (PLA)

PLA is one of the most popular and well-studied biodegradable polyesters due to the large-scale availability from biomass resources, efficient polymerization with ROP, excellent tensile strength and synthetic versatility [80–83]. PLA also exhibits stereoisomerism from the fact that lactide comes in D-, L- and the racemic DL-forms [28,84,85]. This in turn enables the synthesis of isotactic, crystalline PDLA/PLLA and the syndiotatic, amorphous PDLLA [86,87]. The PLLA and PDLA segments can undergo stereocomplex interactions when they are mixed, giving rise to significant enhancement in the mechanical and thermal properties as compared to their parent polymers [88–92]. There has

been extensive studies done both on the use of PLA in SMPs [93] and the exploitation of PLA stereocomplex interactions in various applications [94], but only recently has efforts been devoted into the combination of the two, i.e. making use of PLA stereocomplex interactions to enhance the shape memory performance [95].

Some earlier efforts for biodegradable SMPs focused on incorporating PLA chains to rigid nanoparticle systems which had wellestablished shape-memory properties. For example, polyhedral oligomeric silsesquioxane (POSS) is the smallest possible silica particles that have a well-defined cage-like framework made of silicon and oxygen atoms linked together in a cubic formation [96,97]. It had been previously used in shape-memory polymer designs as physical cross-links in chemically cross-linked systems due to the tendency for POSS particles to crystalize with one another [98,99]. POSS also has the additional advantage of biocompatibility [100]. One representative work was done by Xu and Song [65] who designed and produced an octafunctional POSS-PLA macromer with PLA segments of molecular weights between 5000 and 20,000. The ends of the PLA arms from different nanoparticle units were then linked with bifunctional urethane cross-linkers (Fig. 1A). For comparison, similar systems centred on organic cores in lieu of POSS were also synthesized. Both networks possessed GPa-range room temperature glass-state storage moduli that can rapidly descend to a MPa elastic state at glass transition (Fig. 1B), leading to shape-memory behaviour at similar fixing and recovery temperatures (room temperature and 51 °C respectively); however, the T_g of the POSS-SMP networks was > 10 °C lower, leading to a much faster shape recovery than the organic centred systems at similar temperatures (Fig. 1C). Grid search analysis of the two types of networks revealed that in POSS-SMP networks, the PLA arms were distributed more homogeneously around the bulkier POSS core and therefore had less excessive chain entanglement compared to organic-centred networks, thus lowering the T_g and allowing more PLA chains to participate in shape memory process [65]. The elastic state storage moduli of the networks could be easily adjusted by varying the PLA arm length.

A common drawback of SMPs triggered by external heating is the reduction of mechanical strengths by excessive heat at temperatures above the transition temperature. More recent efforts have been directed at circumventing this drawback by utilizing other mechanisms such as Joule heating of conductive polymers to provide the stimulus for shape transition [101]. For instance, Qi et al. [66] designed a system



Scheme 1. Schematic diagram showing the overview structure of the content discussed in this review.

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