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Featured Letter

Click polymerizations: Encouraging route for shape memory polymers



M. Ragin Ramdas, K.S. Santhosh Kumar*, C.P. Reghunadhan Nair

Polymers and Special Chemicals Division, Vikram Sarabhai Space Centre, Trivandrum 695022, India

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ABSTRACT

Shape memory polymers (SMPs) exhibit exceptional capability to memorise their original shape from a temporary shape by responding to a suitable external stimuli such as heat, pH, current, moisture and so on. Of late, click polymerizations are being emerged as a promising synthetic route to realize novel SMPs. This short-review discusses the recent growth observed in the field of SMPs based on copper catalysed azide-alkyne, thiol-ene and Diels-Alder polymerization methods. The merits, challenges and future directions are addressed.

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1. Introduction

Shape memory polymers (SMPs) are known for their stimuli-sensitive characteristics which enable them to recover original shape from a temporary shape. They display distinctive ability to respond to external stimuli such as heat, pH, moisture, light and electric/magnetic fields [1–4]. As a smart material, they find use as stents, sutures, artificial joints, scaffolds in biomedical field; self-adjustable apparels in textiles; and as self-deployable antennae in aerospace arena [5–8]. SMPs triggered by heat as stimulus are the most common and the key address is their activation temperature (T_{act}) which can be glass transition temperature (T_g), melting transition (T_m) or weak bonds such as hydrogen bonds (supramolecular SMPs). A general classification of SMPs based on their nature of switching unit can be Type I-thermoset SMPs (T_g based), Type II- thermoplastic SMPs (T_m based) and Type III-supramolecular SMPs (H-bond based) (Fig. 1). A macromolecular architecture containing a permanent cross-link (chemical/physical) and a switching segment can exhibit shape memory properties [9,10]. The shape memory potential of an SMP is characterised by two parameters viz: extent of shape fixity (S_F %) and shape recovery (S_R %) which can be quantified generally by fold-deploy test. Programming (testing) of shape memory comprises four stages (i) heating above T_{act} , (ii) application of force to obtain a temporary shape, (iii) fixing of temporary shape by lowering the temperature to $< T_{act}$ and (iv) recovery of original shape from fixed shape by

heating above T_{act} . During the shape fixing process, stress or strain energy gets stored in the polymer chains and kinetically trapped (entropy is locked). The entropy trapped in polymer chains is released on applying an external stimulus which facilitates the recovery of original shape [11].

In the past, popular chemistries such as epoxy-amine, isocyanate-hydroxyl, radical polymerizations (e.g. SMP from polystyrene) and epoxy-cyanate reactions were employed in quantum to achieve variety of SMPs [12–14]. These routes are highly successful in realizing thermoplastic and low T_{act} SMPs which are useful mainly for biomedical applications. Currently, applications of SMPs are extended to high-tech areas like aerospace where high T_{act} , high mechanical properties and low process temperature are indispensable [15–17]. Thermoset SMPs are the suitable candidates (compared to thermoplastic) for these kind of smart-*cum*-structural applications where durability and high strength are also a matter.

Along with the existing routes, click polymerizations have featured a more optimistic sign in the field of SMPs in its entry itself. Click chemistry, introduced by B. Sharpless and co-workers in 2001, comprises organic heteroatom coupling reactions that follow a set of conditions. Click chemistry deals with a group of reactions which can satisfy the conditions such as rapid kinetics, efficiency, versatility, high functional group tolerance, quantitative conversion and no residual by-products. Generally, these reactions can be carried out at relatively milder experimental conditions. [18]. Diels-Alder, thiol-ene and azide-alkyne cycloaddition reactions are the common click reactions broadly employed in polymer chemistry for synthesising functional polymers, new macromolecular architectures, well defined polymers by combining

* Corresponding author.

E-mail address: santhoshkshankar@yahoo.com (K.S.S. Kumar).

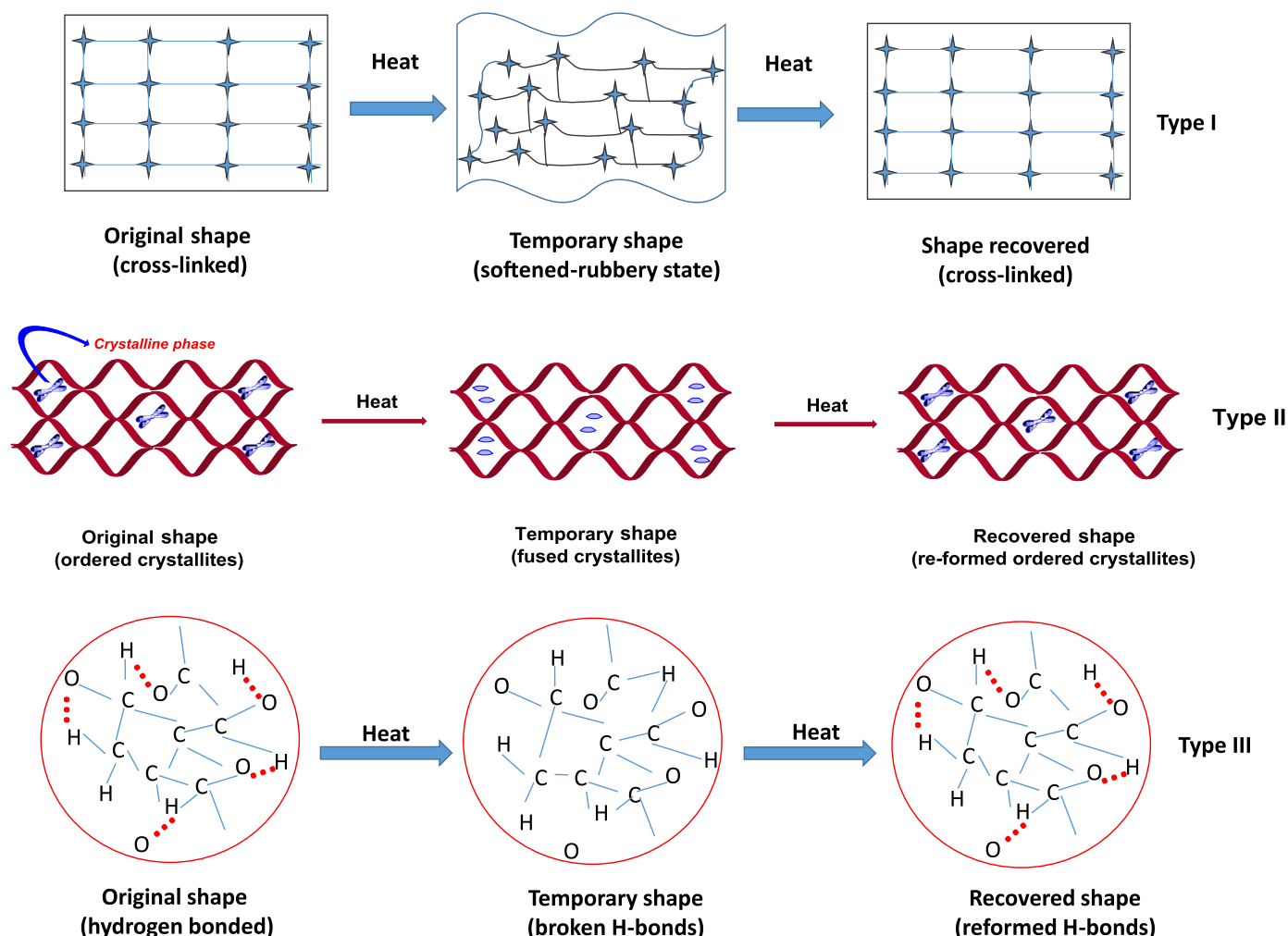


Fig. 1. A general classification of SMPs based on their origin of switching unit; change in macromolecular organization on shape fixing and recovery stages.

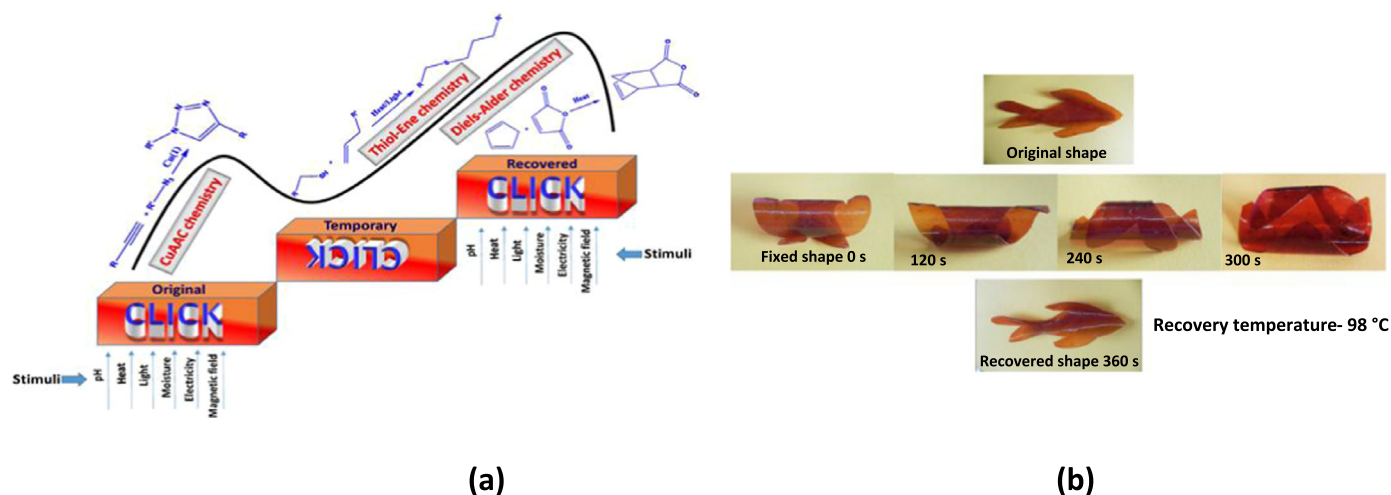


Fig. 2. (a) Typical click polymerizations and schematic representation of shape memory effect (b) shape recovery of an SMP thermoset derived from CuAAC reaction.

atom transfer radical polymerizations (click-ATRP), end-functionalised polymers, smart polymers, surface functionalized polymers, and for coupling polymers [19–27]. Recently, click chemistry is started to utilize for the synthesis of SMPs and this field has witnessed a rapid growth. Rather than coupling different polymers, click monomers are capable to react at moderate polymerization temperature to form cross-linked SMPs. To cite, we

reported an SMP with T_{act} of 123 °C using copper catalysed azide-alkyne polycycloaddition (CuAAC) where reaction temperature was only about 140 °C [28]. Fig. 2a shows basic click reactions adopted in SMP field and a cartoon of shape memory effect. Fig. 2b demonstrates the shape recovery of an SMP thermoset derived from CuAAC polycycloaddition reaction ($T_{act}=73$ °C). The well-known Diels-Alder chemistry was the early one utilised for

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