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Review Article

Review of Memory Polymeric Fibres and Its Potential Applications

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Abstract

Shape memory polymers have attracted a compelling research interest from both academic and industry researchers due to its fascinating behavior. Fibers are fine substances having high ratio of length to thickness and they can be spun using segmented Memory Polymers (MPs). The significance of this review is on the research in memory polymeric fibres in terms of effect of spinning methods, mechanical and cyclic tensile properties, and post treatment processes on their performance. We also discuss the application of Memory Polymer and Fibres (MPFs) into textiles and its characterization techniques. In addition, we highlight our recent discovery of novel stress memory phenomenon in MPs and its implication into smart compression stockings. MPFs are potential to serve in different arenas such as pressure garments, artificial muscles, smart filtration, drug-controlled release, antibacterial nanomaterial, wound dressing, biodegradable sutures and scaffolds for bone tissue engineering, orthodontics, and vibration damping structures. This review also draws conclusions about drawbacks of spinning method, property difference, and recent advances in MPFs and impact of stress memory concept in future.

Keywords: Memory polymer; Fibres; Stress memory; Compression stocking; Textiles

Abbreviations

MPs: Memory Polymers; MPU: Memory Polyurethane; MPF: Memory Polymeric Fibres; SMA: Shape Memory Alloys; T_{trans}: Transition Temperature; T_g: Glass Transition; T_m: Melting Transition.

Introduction

Shape Memory Polymers (SMPs) are important class of stimuli responsive smart materials and have been gained a significant research interest over past few decades [1-5]. The term “shape-memory” was first proposed by Vernon in the year 1941 [6]. SMPs can undergo significant macroscopic deformation and they can be programmed to one or many shapes which recovers back spontaneously to its permanent conformation upon exposure to an external stimulus such as heat [7], light [8], electricity and magnetism [9-11]. SMPs are not only limited to store the shape, it also provides the privilege to memorize and retrieve other physical properties such as stress [12] (stress-memory), temperature [13] (temperature-memory), chrome [14] (chrome-memory) and electricity [15] (electric memory). Hence, they could be termed as Memory Polymers (MPs). Fibres are fine substances with a high ratio of length to thickness and they can be produced with several polymer systems to exhibit interesting memory behaviors. Practically all polymerization methods can be followed to synthesize MPs. The critical/switch temperature of polyurethanes composed of two-phase heterogeneous structure is easily controllable and synthesized via Bulk or Solution polymerization techniques. Segmented polyurethane based MPFs are composed of thermodynamically immiscible fixed and reversible alternative phases and they can be spun via melt, wet, reaction, dry, and electro spinning methods with tunable functionality for proper applications. They have either glass or melting transition for reversible phase. In general, fibres of cylindrical, hollow, nano, and electro active composite can be produced with variable diameters/linear densities with functional properties.

Currently there are several review papers available focusing the polymers/composites for vivid applications. This paper aims to review the memory polymeric fibres in terms of preparation methods, effects of polymer composition/post treatments thermal, mechanical, and cyclic tensile properties, and method of spinning. Interestingly, a novel phenomenon of “stress memory” which was recently discovered in segmented MPs and having great potential to serve where stimuli responsive stress is needed. In addition, implications of stress memory concept in smart compression management and role of MPUs/MPFs in textiles will also be discussed here.

Memory Polymers and Its Molecular Mechanism

Scientific communities are much interested in discovering new phenomenon in the arena of memory polymers. Memory polymers having several advantages compared to SMAs [16]: low density & low cost. (MP = 1.25 g/cc, NiTi SMA = 6.4 g/cc), easy processing with high quality film/foams/wires, extremely high recoverable strain than SMAs. (100% to 95% in solids and foams respectively than 10% with SMAs). In addition, the thermo-mechanical properties can be tuned by blending with different types of fillers with wide range of shape recovery temperature (from -20^{0}C to +150^{0}C). They provide excellent chemical stability, biocompatibility and biodegradability in responsive to multiple stimulus. MPs are intrinsically sensitive to ambient temperature and responsive to narrow range of temperature (Figure 1a).

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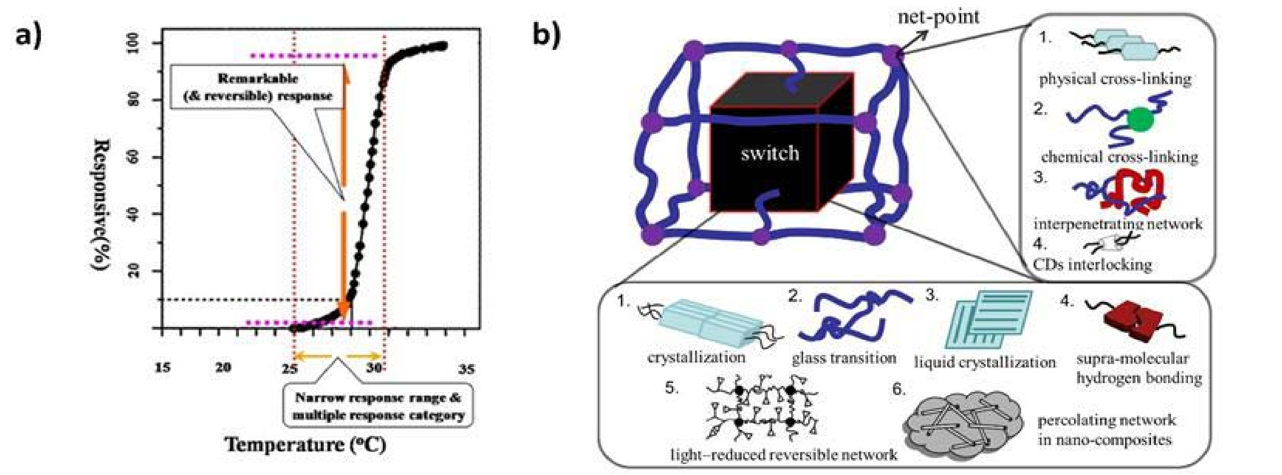

Figure 1: Memory polymeric architecture and its mechanism.a) Plot depicting the sensitive change of MP in ambient temperature; b) The overall architecture of MPs [16].

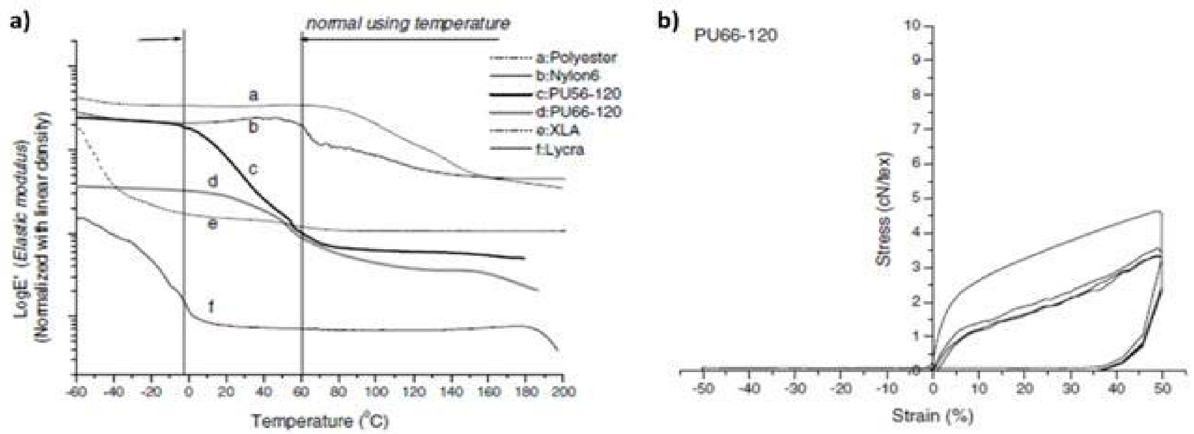

Figure 2: (a) Comparison of elastic modulus between MPFs and various manmade fibres. (b) MPF showing 100% shape recovery.

Hu et al, proposed an overall 3D MP architecture (Figure. 1b) based on the molecular mechanisms [1]. This model can describe the structure of any MP and it consists of both switch units and net-points. The net-points (hard segment) determine the permanent shape and can be made of either chemical or physical cross-links, with an interpenetrated or interlocked supramolecular complex. The driving force for strain recovery in MPs is the entropic elasticity of the polymer network. Basically, switch unit is the soft segment and responsible for shape fixation upon deformation and recovery upon certain stimulus. The switches such as amorphous, crystalline and LC phases, supramolecular entities, light-reversible coupling groups and newly utilized percolating cellulose-whisker networks have served as soft segment in controlling the SME of SMPs.

Transition Types of MPs

Transition is the temperature range of a polymeric system where the significant change in modulus and shape occurs due to change in temperature. Basically SMPs has glass (T_g) or melting (T_m) type of transition for the soft switch segment. The network chains of the soft segment in MPs can be either crystalline or amorphous therefore the T_{trans} can be either melting or glass type respectively [4,17,18]. If the $T_{trans} = T_m$, strain induced crystallization [19] occurs in the material upon deforming and cooling below the T_m. The shape recovery of the SMPs are prevented by the crystallites and until it is reheated above T_m [4]. Tobushi and Takahashi et al. [20,21] stated that, if the $T_{trans} = T_g$, the micro-brown motions of the polymer network will be frozen and set into glassy state when cooled below T_g. The network will be in non-equilibrium state until it is reheated above T_g to activate the micro-brown motions. There are different compositions of hard and soft segments available and which decides the transition type [22-32].

Memory Polymeric Fibres (MPFs)

Memory Polyurethanes (MPUs) are synthesized via solution or bulk polymerization techniques. Polydiol is used as a soft segment and isocyanate and molecular extenders are as hard segment content to synthesize MPUs. The MPUs can be spun into fibres via melt spinning, wet spinning, dry spinning, reaction spinning, and electro spinning methods with additional functionalities to prepare them for specific applications [33].

Hu et al. have developed MPU filaments (MPFs) using polyol

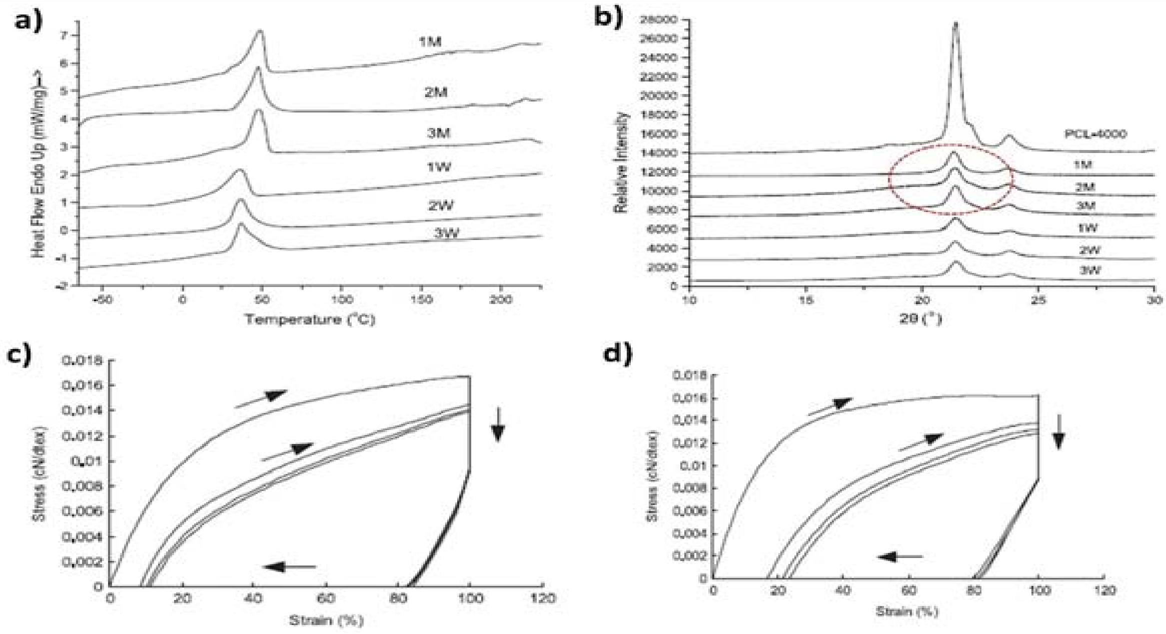

Figure 3: Influence of Spinning methods on thermal and shape memory properties. (a) DSC thermographs of melt and wet spun MPFs. (b) XRD profiles of melt and wet spun MPFs. (c &d) Stress-strain curves of melt and wet spun MPFs.

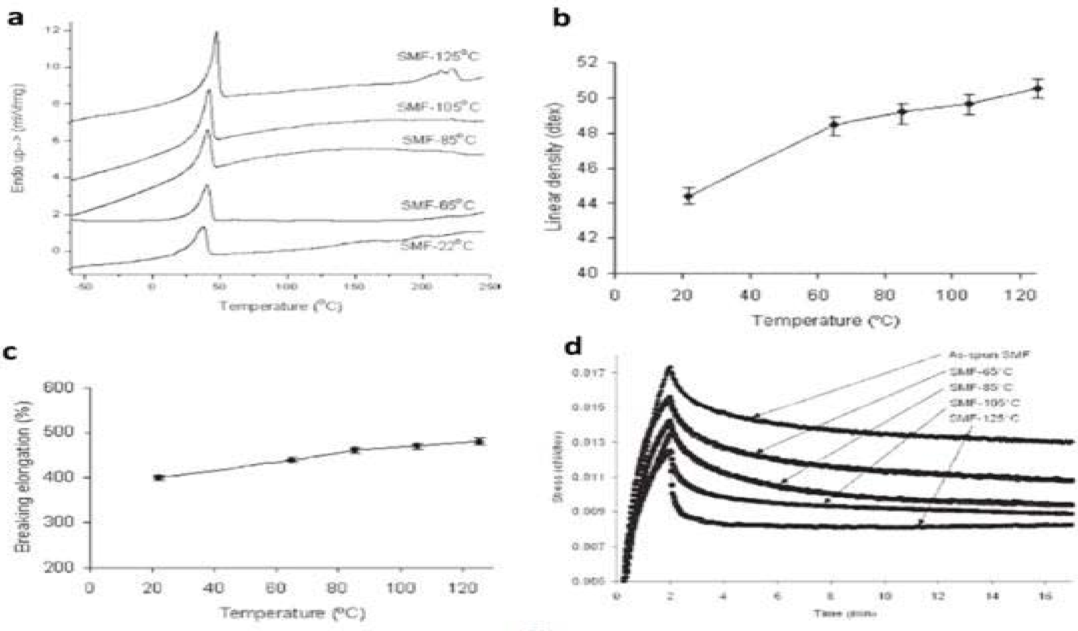

Figure 4: Influence of heat treatment on properties of as spun and heat treated MPFs. (a) DSC thermograms of as spun and heat treated MPFs. (b) Change in MPF linear density. (c) Change in MPF breaking elongation. (d) Effect on stress relaxation.

as soft segment and small size polydiols/MDI as hard segment by different spinning techniques; wet, reaction, dry, melt, and electro spinning [33,34]. A systematical investigation has been carried out by Hu et al in 2006, to identify the relationship between the thermal, mechanical, and shape memory properties of MPU filament and other man-made fibres (Figure. 2a). They also synthesized MPFs with different hard segment content via pre-polymerization method and spun by wet spinning process to achieve complete shape recoverability [26].

Meng et al have conducted an extensive research to find out the relationship between morphology, phase separation, thermal and mechanical properties of filaments prepared by different spinning methods and understood the underlying physics behind the property differences. It was shown that MPFs prepared by melt spinning method having high tenacity, breaking elongation, and high degree of phase separation [35]. Figure 3a shows that, 1,2, 3M MPFs have

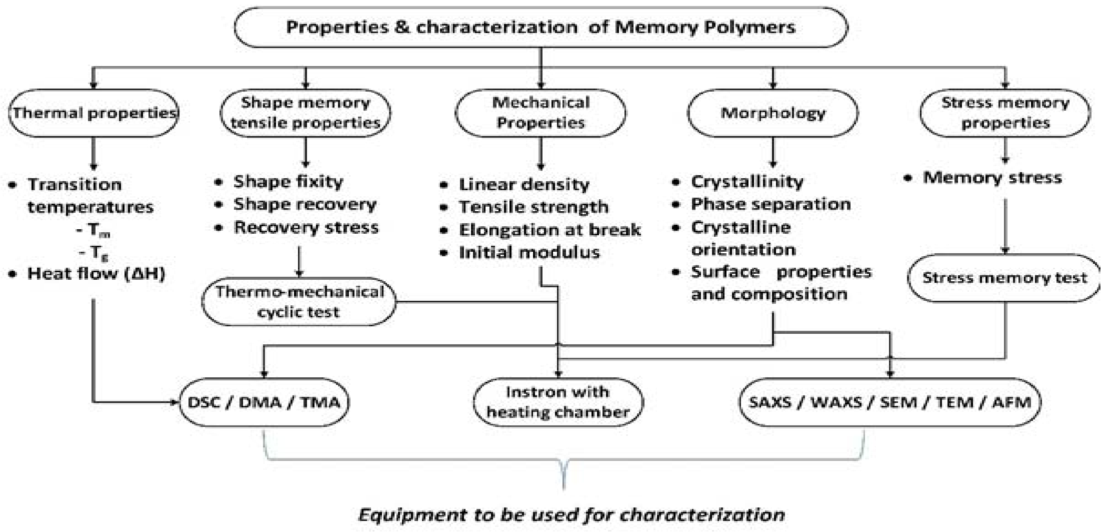

Figure 5: Characterization techniques of memory polymers/fibres.

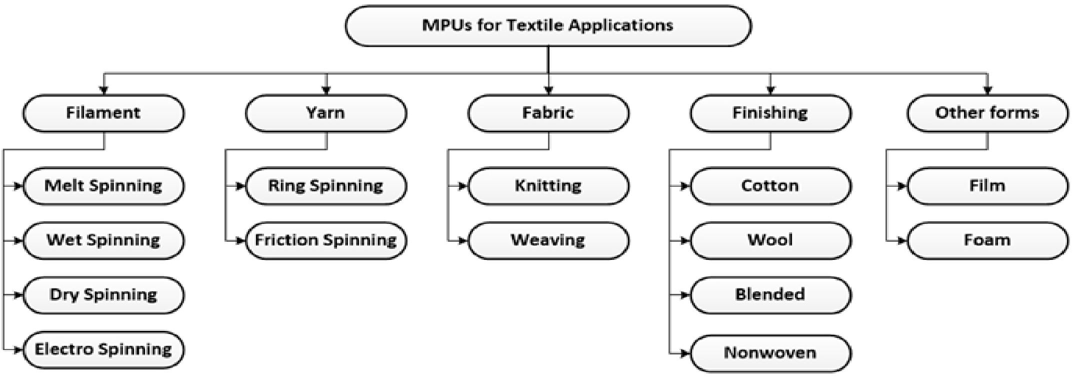

Figure 6: Different forms of MPs for textile applications.

higher T_m ,crystallinity, significant soft and hard segment transition with relatively good hard segment rich phase than wet spun MPFs. The XRD profiles (Figure. 3b) of melt spun MPFs also shown more prominent peaks of soft and hard segment phases of melt spun MPFs in contrast to wet spun MPFs. Melt spun MPFs (shape fixity-86%, shape recovery-98%) were resulted to have a very good shape memory properties than wet spun MPFs (shape fixity-82%, shape recovery-95%) (Figure. 3c,d). Smart shape memory polyurethane hollow fibre was prepared by Hu et al. via melt spinning and which is able to change and recover the diameter under thermal stimulation. The hollow fibre showed tenacity of 1.14 cN/dtex, breaking elongation of 682%, and shape fixity of 87% with 89% recovery ratio. These could be used for the applications such as smart filtration, drug-controlled release and in other suitable fields [36].

Electro Responsive MPU Fibres

Electro responsive MPU fibres were developed by Hu et al. and which could recover their shapes under electrical stimulation. The Shape Memory Polyurethane (SMP–MWNT) composite was prepared by in situ polymerization and the MPU–MWNT fibre was prepared by melt spinning. CNTs were incorporated into the MPU fibres and which are charged at the both the ends. Voltage required to generate the heat to trigger the shape recovery was still high. Scanning electron microscopy and Transmission Electron Microscopy (TEM) observations of the morphology revealed that the MWNTs are axially aligned and homogenously distributed in the MPU matrix, which is helpful for the fibre's electrical conductivity improvement and for the electro-active shape memory effect [37].

Influence of Thermal Setting/Heat Treatment

Heat treatment /thermal setting process is necessary in order to eliminate the internal stress or structure deficiency of the filaments caused by spinning process. Thermal setting temperature has a significant influence on the shape memory and mechanical properties of the MPFs and this has been investigated by Zhu et al [26]. It has been shown that, thermal setting with a higher temperature will result in lower tensile modulus, tenacity, and higher elongation at break. With an application of optimal heat setting treatment, complete shape recoverability can be achieved in MPFs due to counteracting effect of irreversible strain and thermal shrinkage.

Figure 4a shows that increase in the temperature of heat treatment results in higher soft segment crystallinity due to reduced chain entanglement and also ordered hard segment content. This shows the ability of the heat treatment to repair the destroyed crystals in PU. It was also reported that prominent increase in the linear density and breaking elongation of MPFs due to release of stored internal stress and molecular disorientation at higher temperature (Figure 4b,c). Figure 4d show the stress relaxation of MPFs with time, higher the temperature results in greater the stress decay and reaches the plateau with more stability [38].

Effect of Drawing

Meng et al., have studied the effect of thermal and cold drawing on the shape memory properties of MPFs via cyclic tensile testing in comparison with commercial polyurethane fibres [39]. Thermal and cold drawing refers to temperature maintained during the cyclic tensile testing process. It was reported that the modulus of MPFs was higher than commercial fibres in the ambient temperature and had a shape fixity and recovery ratios of more than 80% and 95% under thermal drawing and recovery respectively.

Shape Memory Yarns

MPFs can also be used in the form of yarns covered with cotton fibres. Hu et al. have developed SMP yarns using polyurethane SMF as a core and cotton fibres as a sheath by ring and friction spinning methods [40]. It was reported that ring spun core yarns were having higher tenacity compared to friction spun yarns and breaking elongation of core spun yarns were lesser than MPFs. The shape fixity of ring spun and friction spun core yarns were 97.5% and 94.5% respectively. These kind of yarns could find the applications in protective textiles, functional textiles, apparels, etc.

Characterization Techniques for Memory Polymers/Fibres

Figure 5 shows the different techniques and equipment to be used to characterize the structural, physical properties and its relationship with the polymeric system. The techniques tabulated here is applicable for polymers in the form of bulk, film, filament, yarn, and foam.

Applications of Thermal-induced MPUs into Textiles

MPUs are basically stimuli responsive polymers and they can be applied to textiles to enhance the smart functions in 2 ways: Finishing or built-in methods. Finishing process involves basically a coating or laminating techniques. While built-in method includes the blending and spinning. SMPs can be applied on to textiles in different forms such as emulsion, solution, film, fiber, foam, and bulk forms under specific conditions. Application of MPU into textiles provides several advantages; 1) The switching temp can be tailored even to set around body temperature, 2) Superior processibility, 3) Soft mechanical properties, 4) High strain deformability and recoverability.

Recent Advances in the Field of MPFs

Researchers have been continuously working in the field of memory polymeric fibres to implement them into multidisciplinary areas such as biomimetic fibrous scaffolds, fibrous structures for vibration damping, fiber supercapacitors for energy harvesting, fiber assembly for artificial muscles, and fiber based composites for healing applications. Zhang et al. have fabricated a biodegradable Nano fibrous scaffolds with shape memory properties and implied into bone tissue engineering offering shape fixity and recovery ratios more than 90% [41]. MPFs based supercapacitors offers excellent stability in electrochemical performance suitable for programming and recovery from temporary shapes [42,43]. Li et al. have proven that MPFs have higher toughness (276-289 MJ m^{-3}) compared to spider dragline silks (160 MJ m^{-3}) due to excellent stretchability and having higher vibration damping capability [44,45]. Li et al. have also developed hierarchically chiral structured artificial muscles based on two-way shape memory fibres. Experimental results have shown that the negative coefficient of thermal expansion is one order higher than those made of polyethylene fibres [46]. MP fibres have great potentials and would provide a broad horizon to tap other applications in near future.

Implication of Memory Fibres into Smart Compression Stockings

Gravity is the main reason why chronic disorders occur in the lower extremity of the human body [47]. Compression therapy is considered as a cornerstone in the conservative treatment of phlebological and lymphatic diseases such as venous leg ulceration, varicose veins, venous hypertension, venous oedema, venous stasis, Deep Vein Thrombosis (DVTs), and other chronic venous disorders [48-52]. The principle objective of compression therapy is to apply certain level of pressure around the affected area [53-55]. Maintaining proper level of pressure in the stockings has always been a contest for both clinicians and practitioners due to several attributes. In line with this, Hu et al., have recently discovered a novel phenomenon of "stress-memory" in memory polymeric system and applied into stockings to resolve the conventional problems [12,56]. The smart compression system offers multi-functionalities such as massage benefits, selective pressure control, size fitting.

Conclusions

Memory polymers have several advantages compared to alloys such as low density, low cost, and easy processing. MPUs are mainly synthesized via solution or bulk polymerization depending on the method of fiber spinning is chosen. The different composition of hard and soft segment contents decides the type of transition (T_m or T_g). The spinning methods such as melt spinning, dry spinning, wet spinning, reaction spinning, and electrospinning can be followed to spin the fibres with desirable linear densities or type of cores with additive functionalities. Melt spinning is desirable to produce fibres with high tenacity, breaking elongation, crystallinity, shape fixity, shape recovery, and high degree of phase separation compared to wet spinning. Thermal setting at high temperature results in lower tensile modulus and tenacity with higher breaking elongation, whereas optimal treatment helps to achieve complete shape recoverability. Drawing has a significant effect on fibres in achieving high shape fixity and recovery ratios. MPUs offers different platforms in the forms of filament, yarn, fabric, finishing solutions, and films/foams to apply into textiles for numerous applications.Hu et al. have also developed MPFs such as thermal sensitive hollow fibres and electro responsive fibres which could be used for the applications such as smart filtration, drug-controlled release, antibacterial nanomaterial, wound dressing, biodegradable sutures and scaffolds for bone tissue engineering, orthodontics, fiber supercapacitors, artificial muscles, and vibration damping structures. In addition, the novel "stress-memory" concept which was recently discovered by Hu et al. has a great potential to imply into smart compression stockings to provide multi-functional benefits such as massage effect, selective pressure control, and size fitting. This unprecedented approach would have a great impact in the smart compression management in upcoming days.

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