Feasibility study of polyurethane shape-memory polymer actuators for pressure bandage application

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Received 23 August 2011
Accepted for publication 3 January 2012
Published 2 February 2012
Online at stacks.iop.org/STAM/13/015006

Abstract

The feasibility of laboratory-synthesized polyurethane-based shape-memory polymer (SMPU) actuators has been investigated for possible application in medical pressure bandages where gradient pressure is required between the ankle and the knee for treatment of leg ulcers. In this study, using heat as the stimulant, SMPU strip actuators have been subjected to gradual and cyclic stresses; their recovery force, reproducibility and reusability have been monitored with respect to changes in temperature and circumference of a model leg, and the stress relaxation at various temperatures has been investigated. The findings suggest that SMPU actuators can be used for the development of the next generation of pressure bandages.

Keywords: advanced shape-memory polymer, recovery stress, recovery force, compression bandages, venous leg ulcer

1. Introduction

Shape-memory polymers (SMPs) are an emerging class of polymers, which have the ability to change from their temporary deformed shapes to their permanent shapes upon stimulation, and generate a recovery force through the release of the deformation energy simultaneously. They have the advantages of large strain, easy processing, low cost and possible biocompatibility. Owing to their novel and excellent properties, they can be used in a broad range of applications [1,2], and the development of these materials is often motivated by the requirement of specific applications such as smart textiles, aerospace industry and medical devices [3,4]. Although SMPs possess low mechanical strength and recovery stresses compared to shape-memory alloys, high deformation and shape recovery ratios make them suitable for various applications, especially for those where high recovery stress (force) is not essential [5].

In this work we have explored the feasibility of using a specially synthesized polyurethane-based shape-memory polymer for possible pressure bandage application by investigating fundamental behaviour and functional properties of these advanced materials under different testing conditions.

One in ten adults aged above 50 years in UK suffers from venous leg ulcer [6]. Its treatment costs UK hundreds of millions pounds [7]. In this disease, the lower leg deteriorates due to low physical activity and poor blood circulation, especially for the elderly patients. For patients with venous disease, the application of graduated external compression can help to minimize or reverse the vascular changes; this is normally achieved by forcing blood from the interstitial spaces back into the vascular and lymphatic compartments. For this reason it is usual to ensure that external compression is applied in a graduated fashion, with the highest pressure at the ankle and lower pressure at the upper part of the leg. In practice, the optimum pressure will probably vary according to a number of factors, including the severity of the condition and the height and limb size of the patient [8].

Various types of commercial bandages are available in the market for venous ulcer treatment such as lightweight conforming-stretch bandages; these bandages have good elasticity, but produce low compression forces. Similarly,
light support bandages impart good compression but have poor elasticity. Medical hosiery is a useful and convenient method of applying a graduated compression to prevent the development or recurrence of leg ulcers. Compression bandages currently represent the treatment of choice [9, 10]. However, hosiery and compression bandages are of limited value in the treatment of active ulceration, difficult to apply over dressings, tend to lose mechanical strength and stiffness due to breakdown of fibres involved [11], and are expensive [12]. Similarly, slippage or displacement in compression bandages results in the formation of multilayers, which lead to high pressures on affected area, deteriorating the ulceration. These bandages also lose the pressure gradually as time passes due to loss of mechanical strength of the fabrics. Air-bag type bandage is another type of dynamic bandage [13, 14]: it is limited to use in hospitals under supervision of experienced nurses, and cost of this treatment is rather high [15, 16].

Specially synthesized polyurethane shape-memory polymer (SMPU) strips that are pre-stretched under heat may offer an alternative means of this treatment. A series SMPU strips will generate the required pressure gradient via shrinking forces when stimulated by a heat source, and this property may be useful for venous leg treatments [17]. We have conducted the feasibility study of the SMPU-related pressure actuators, and report on fundamental behaviour of laboratory-produced SMPU actuators when exposed to known temperature settings, particularly on the induced pressure under different circumstances.

2. Design and test experiments

2.1. Design principle of the envisaged bandage

Figure 1 shows two designs of the proposed SMP bandages consisting of strips with fixed strain of different lengths or fixed length of different strains [17]. Temperature-responsive SMP strips are attached to fabrics which will be part of the underlay padding of bandages. For easy actuation, SMP with low transition temperature, around \( \sim 50 \, ^\circ\text{C} \), will be used. The SMP strips are pre-stretched to fixed lengths or strains. In its fully developed form, the SMP bandage would be wrapped on the venous leg and held in position by Velcro fasteners or other similar means. Upon application of an external heat using a hair drier or a hot towel, the SMP stripes will shrink and partially return to their original lengths, providing pre-defined forces or pressure distribution for venous leg treatment. The pressure, \( P \), with coaxial forces acting inwards by the shrinking force, \( F_s \), of the SMP strip as shown in figure 2(b) can be estimated as

\[
P = F_s/\text{area} = F_s/(WL_g).
\]

The area is the product of the SMP strip width, \( W \), and the circumference of the limb, \( L_g \) (note this is different from the length of the stretched SMP strip), which is a function of the position of the limb as shown in figure 2(c). \( L_g \) is small near the ankle and increases as it approaches the upper leg. A typical peripheral length of a limb is \( \sim 25 \, \text{cm} \) near ankle, and increases to \( \sim 50 \, \text{cm} \) at the upper limb [18], depending on person. Therefore, when SMP/fabric bandage with the pre-set pressure distribution is used with the structure shown in figure 1, a gradient pressure along the bandage can be achieved, which could be suitable for leg ulcer treatments. For the ulcer leg treatments, the typical pressure required at the ankle is 40 mmHg, and decreases to 15 mmHg at the upper part of the leg as schematically shown in figure 2(c). For easy understanding of the bandage development, the pressure unit of mmHg is used for the pressure generated by the SMP bandages hereafter, rather than the Pascal which is generally preferred in materials research (1 mmHg = 1 Torr \( \approx 133 \, \text{Pa} \)).

If the pressure drops during use, it can be readjusted using a hair drier or a hot towel to warm up the SMP bandage, locally or as a whole. This can be done by the patient at home without a need for an experienced nurse, significantly simplifying the treatment. Furthermore, the SMP strips are only used as the pressure provider, not the medication; therefore, they can be reused or recycled many times, significantly reducing the cost of the treatment.

2.2. Materials used and test methodology

In the following sections, we present tests of SMP actuators to clarify whether they are able to meet the basic requests. For this purpose, polyurethane-based SMPU has been chosen for the pressure actuator experiments owing to its excellent thermal stability, easy synthesis and device fabrication processes, and low cost. Recently we have developed a process to synthesize SMPUs with tailored transition temperatures, \( T_g \), in the range of 30–60 \( ^\circ\text{C} \) using various polyols as soft segments and a combination of 4,4'-diphenylmethane diisocyanate (MDI) and isophorone

![Figure 1. Two designs of the proposed SMP/fabric bandages.](image1)

![Figure 2. An SMP strip fixed on a fabric and direction of the force generated once the strip is activated (a), the co-axial force can be generated by wrapping the SMP strip on a circular shape once activated (b); (c) is a model leg showing a varying circumference and pressure required for the leg ulcer treatment.](image2)
corresponds to the sample PCL-PU-2 in 11.5% and BDO concentration of 2.5%. This PCL-SMPU has a molar concentration ratio of 6.24 MPa, suitable for the proposed investigation. This PCL-based SMPU consists of 58% soft segment and 42% hard segment. The latter has a molar concentration ratio of MDI/(MDI + IPDI) = 0.68, PEG-200 concentration of 11.5% and BDO concentration of 2.5%. This PCL-SMPU corresponds to the sample PCL-PU-2 in [19].

The feasibility of the SMP bandage was evaluated on example of SMPU synthesized with polycaprolactone (PCL-2000) as the soft segment. It has a transition temperature of 45 °C, good flexibility (maximum strain over 800%), a recovery ratio of 82% and stress at break of 6.24 MPa, suitable for the proposed investigation. This PCL-based SMPU consists of 58% soft segment and 42% hard segment. The latter has a molar concentration ratio of MDI/(MDI + IPDI) = 0.68, PEG-200 concentration of 11.5% and BDO concentration of 2.5%. This PCL-SMPU corresponds to the sample PCL-PU-2 in [19].

The synthesis was carried out as follows: a three-necked flask was immersed in a water bath, which was purged with a continuous flow of dry nitrogen and equipped with a mechanical stirrer and a thermometer to monitor the temperature. A desired quantity of the polyol was firstly added into the flask containing N,N-dimethylformamide solvent, followed by adding a required amount of IPDI. The chemicals were allowed to react for 2 h at 90 °C, then PEG-200 was added into the reaction flask, followed by adding MDI in the flask. The mixture was stirred for 1 h at 90 °C. Two drops of dibutyltin dilaurate were then added as a catalyst, and finally the required amount of the chain extender was added and mixed by stirring for 1 h at 60 °C. To cast an SMPU film, the synthesized SMPU solution was poured onto a PTFE-coated glass mould and baked at 60 °C for 12 h, then at 80 °C for 24 h, and further at 100 °C for 8 h, all in a vacuum oven. The average film thickness was 0.60 ± 0.05 mm after drying.

The films were cut into 70 × 30 mm strips. Each strip was stretched to a pre-defined strain of 25, 50 and 75% in the heated chamber of Instron 3369 tester and cooled down to room temperature. After sufficient time (at equilibrium state), free ends of each strip were attached to a piece of knitted fabric using glue (Araldite Rapid) to allow gripping of the SMPU samples without affecting the force generated by SMPU strip. Only the middle 50 mm of an SMPU strip was used for extension due to the limited gauge length and the use of 10 mm on each end for clamping.

For accurate measurement of uniform pressure generated by the SMPU actuator, glass cylinders with a fixed diameter of different circumferences (L_f = 90, 100 and 125 mm) were used as the model leg. The prepared SMPU strips were, one at a time, wrapped around the cylinders and held in place via suitable clamps before being put in a temperature-controlled oven (EC-2007, Severn Thermal Solution UK). The assembly was heated at a rate of 10 °C min⁻¹ for the investigations at different temperatures and strains. The pressure generated by the SMPU actuator was monitored using a pressure sensor (Kikuhime™, a sub-bandage pressure monitor) placed between the cylinder surface and the actuator strips. The SMPU strips were tested three times at the same strain up to the same temperature to verify the repeatability and reliability of the generated pressure and pressure adjustment by repeated heating, and the average values with typical errors of less than 15% were recorded.

3. Results and discussion

3.1. Relationship between pressure and strain

Figures 3(a) and (b) show the stress–strain relationship and the pressure generated with temperature for an SMPU strip. The sample was firstly extended to 25% and cooled to room temperature (T_g ∼ 25 °C), and it was then wrapped on the cylinder for pressure generation experiment by ramping the temperature up to 60 °C, sufficiently higher than the T_g of the SMPU to ensure it is fully activated, and down to T_r (figure 3(b)). The same actuator was subjected to repeated experiments for 5 times (N-1 to N-5) and the results are summarized in figures 3(a) and (b). The large drop of the stress from the first test to the second test is mostly caused by internal molecular rearrangement and chain disentanglements in the first round of extension, and is a typical characteristic of polymers [20, 21]. Subsequent stress–strain curves are much

Figure 3. Stress of an SMPU strip as a function of strain up to 25% extension (a) and the pressure generated by the strip (b) as a function of temperature. Both were measured five times to test for repeatability and reproducibility.
**Figure 4.** Maximum pressures generated by an SMPU strip at 60 °C (a) and 25 °C (b) as a function of strain and number of test cycles.

In the first heating round, the pressure generated by the SMPU strip gradually increases with temperature and reaches the maximum level of 25 mmHg at 60 °C. Upon cooling, the pressure drops from 25 to 14 mmHg at 25 °C. In subsequent cycles, the maximum pressure generated at 60 °C is 22, 20, 19 and 18 mmHg for the 2nd to 5th cyclic tests, respectively, and the corresponding pressure at $T_r$ decreases from 14 to 10 mmHg. Both the pressures at 60 °C and $T_r$ continuously decrease with the increase of the test number which is attributed to the cumulated disentanglements of molecular chains and bonds by the sequential extension. These results imply that the SMPU actuator strips can be readjusted by using higher activation temperatures, and be reused for many times through full recovery at high temperatures as shown in our previous work [19]. The pressure increases nonlinearly with temperature on warming-up and is linearly correlated with temperature on cooling down. The nonlinearity can be attributed to the changes in molecular arrangement resulting from the increased molecular mobility of the polymer, which transitions from its glassy to rubbery state and releases its energy/strain stored during initial deformation. On cooling, however, the phase transition is not coupled with a shape transformation (i.e. no major chain rearrangement occurs) that leads to the linear relationship between pressure and temperature.

**Figure 5.** Dependences of pressure on the strain of an SMPU strip with $L_g = 100$ mm at 60 °C and 25 °C ($T_r$).

**Figure 6.** Relationships between the maximum pressure and circumference obtained at 60 °C and 25 °C ($T_r$).
be expressed by the following equation for both 60 and 25 °C cases:

\[ P = (0.31 \pm 0.03)\varepsilon + P_0. \]  

Here \( P_0 \) is the initial pressure which is related to SMPU properties such as elastic modulus, actuation speed and the initial pressure set at the wrapping stage; the ‘+’ sign is for room temperature, while the ‘−’ sign is for the 60 °C case. This equation implies that the pressure distribution can be achieved by changing the strain of the SMPUs strips used in the bandage.

3.2. Relationship between pressure and circumference

Since pressure is inversely proportional to the area as expressed by equation (1), pressure decrease is expected as the circumference of the leg increases at a fixed length of the SMPU strip. This provides another way to design a distributed pressure bandage. Figure 6 illustrates the pressure variation as a function of circumference at 60 and 25 °C for SMPU strips with 75% strain. The pressure is 46 mmHg for \( L_g = 90 \text{ mm} \) and decreases with the increase of the circumference. The pressure–circumference relationship can be expressed by the following equation:

\[ P = -(0.62 \pm 0.03)L_g + C_1. \]  

Here \( C_1 \) is a constant related to temperature, material properties such as elastic modulus, the design of the device and the initial pressure set at wrapping stage; the ‘+’ sign is for the 60 °C case, while the ‘−’ sign is for the room temperature. These relationships demonstrate that the pressure of the SMPU bandage can be designed and controlled by using different lengths and strains of the strip.

The pressure as a function of circumference was tested repeatedly to verify the repeatability and reusability of the SMPU and the results are shown in figure 7. The maximum pressure decreases with the test number for all the cases. The results show that the SMP actuator can be used for several times, but the pressure (distribution) gradually decreases, and stimulations at higher temperature are needed. The large drop in pressure from the first test to the second tests for both

\[ Figure 7. \] Maximum pressures generated by an SPMU strip at 60 °C (a) and 25 °C (b) as a function of circumference and number of test cycles with a fixed strain of 75%.

\[ Figure 8. \] Pressure drops caused by stress relaxation as a function of time at different temperatures. Fast pressure drop is observed at high temperatures, and it slows down as the temperature is decreased.

\[ Figure 8. \] Pressure drops caused by stress relaxation as a function of time at different temperatures. Fast pressure drop is observed at high temperatures, and it slows down as the temperature is decreased.

3.3. Pressure relaxation

Stress relaxation is one of the typical characteristics of SMPs, as molecule chains may disentangle and oriented chains tend to rotate to align with the direction of the applied force, and it has significant impact on recovery stress (force). The stress relaxation of the SMPU strips will result in pressure decay which would undermine the intended application. Pressure relaxation was measured at different temperatures and is summarized in figure 8. Pressure relaxation is much higher at a temperature above the glass transition temperature, and tends to decrease as the temperature is decreased. The largest drop in pressure was observed at 60 °C from 33 to 4 mmHg after 5 h. As the temperature was lowered, pressure relaxation slowed down, but remained even at 30 °C. A pressure drop by \( \sim 3\% \) after 5 h was observed for the SPMU strip at 30 °C. At a temperature above \( T_g \), molecules became relatively mobile and are more prone to slippage and rotation, leading to...
faster relaxation. Lowering the temperature will reduce the mobility of the molecules and slow down the relaxation. Although there is no significant pressure relaxation at room temperature, slow relaxation at 30–40 °C may bring the concern of pressure drop caused by the body temperature (∼37 °C) and environment. This relaxation can be reduced by using a thick underlay padding and surface layer (both are necessary for products), which will insulate the heating from human body and environment.

For practical use, SMPU with a relatively high \( T_g \) and a sharp transition may be better for prevention of the pressure drop, and such SMPUs are being developed. Furthermore, the pressure drop can be easily overcome by reheating the actuator strips as shown later to gain the pressure required for the treatment. The results have thus demonstrated that the SMP bandages are capable of holding pressures for long time without dropping and can be re-adjusted by heating.

The SMP properties such as the transition temperature and hardness change when immersed in water for tens of hours [22]. As water may deteriorate the SMPU bandage, the SMPU strips have been immersed in water for 2 h at room temperature, and then their thermomechanical properties were tested. This immersion had no measurable effect on the performance of the SMPU actuators, contrary to previous reports, probably due to different materials used for the synthesis of the SMPUs. Therefore, body fluids and sweating should not significantly affect the SMPU performance, at least for a short period, although a more careful investigation would be needed before practical application.

3.4. Pressure readjustment

Inevitably bandage pressures drop during use, as for all compression bandages in the market. However, the advantage of an envisaged SMPU bandage over the existing compression bandages would be that the pressure and its distribution could be easily readjusted by heating the bandage locally or as a whole. To verify the accuracy and repeatability of SMPU strips in this regard, they were subjected to repeated thermal cycle tests up to the same temperature, i.e. 60 °C, without being removed from the cylinder (a situation similar to a bandage in use). The results are summarized in figures 9(a) and (b). Figure 9(a) shows the changes in pressure as the temperature rises while figure 9(b) shows the corresponding pressure drops upon cooling. It was found that the maximum pressure at 60 °C gradually decreased from 32.5 to 31, 30, 29 and 28 mmHg after repeated heating up to five times. The corresponding room temperature pressure decreased as well, implying that repeated heating at the same temperature only leads to decrease of pressure.

It was found that the pressure can be increased when the SMPU strip is heated at a temperature higher than the previous one used. Figure 10 shows the pressure readjustment for an SMPU sample which was strained up to 50% at 60 °C. The strip was initially tested up to 40 °C, and cooled down to room temperature. The pressure generated was 9 mmHg at room temperature. By raising the temperature to 50 °C without removing the strip from the cylinder the pressure was increased continuously. Once the strip was cooled down, the pressure reached a value of ∼13 mmHg at \( T_r \), higher than 9 mmHg of the previous test. Similarly, the pressure reached 22 mmHg at \( T_r \), after up to 60 °C actuation as shown in figure 10. These results clearly show that the final pressure...
at room temperature is determined by the highest stimulation temperature, and the pressure can be readjusted by heating the SMP at a temperature higher than that used in the previous round, demonstrating the reusability of SMP. If necessary, the SMPU bandage can also be unwrapped and re-wrapped to adjust the pressure.

SMPU actuators have been subjected to tests for pressure generation and pressure readjustment at different temperatures and strains. They were also examined for repeatability and reusability and the results were promising for potential use in a new type of SMP-based compression bandage. However, it has to be pointed out that no statistical evaluation, replication of the experiments and safety of the SMPU were considered. The intention of this work has been to illustrate the feasibility of using such smart material systems and to raise the interests for further research. No doubt a lot more research and more realistic models are required to fine-tune such bandaging systems for venous leg applications and for proper design of the bandage with multiple SMPU strips. Our preliminary results suggest that it possible to develop compression bandages using the smart shape-memory polymer actuator strips. These futuristic bandages could drastically reduce costs because of ease of application (and hence elimination of trained nurses) and reusability of these products, i.e. cyclic ability to reshape. However, much more work, especially on the statistics, repeatability and safety, is needed before practical medical trials.

4. Conclusions

This research has shown that a laboratory-developed polyurethane shape-memory polymer can be considered for the development of next-generation medical pressure bandages. It demonstrated four important relationships between temperature and the induced pressure, pressure and the circumference of the test object (i.e. the leg), stress relaxation and the endured pressure decay as well as pressure readjustments. Cyclic tests and tests of bandage pressures at different temperatures have shown the repeatability and reproducibility of these bandages, readjustment of pressure by heating at higher temperature, and potential for multiple reuse.

Acknowledgment

The authors would like to acknowledge the partial financial support received from the Engineering and Physical Sciences Research Council under the grant EP/F06294 and the Leverhulme Trust under the grant F/01431, as well as the Royal Society for the UK-China International Joint Project (RG090609).

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