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# Smart Medical Stocking using Memory Polymer for Chronic Venous Disorders

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## Abstract

Proper level of pressure or compression generated by medical stocking or hosiery is the key element for successful treatment or management of chronic venous disorders such as oedema, leg ulcers, etc. However achieving the recommended compression level and, more importantly, sustaining it using stockings has been a major challenge to the health practitioners supervising the treatment. This work aims to investigate and design a smart compression stocking using shape-memory polymer that allows externally controlling the pressure level in the wrapped position on the leg. Based on thermodynamical rubber theories, we first derived several criteria that have to be satisfied simultaneously in order to achieve the controlled pressure adjustment using external heat stimuli. We then presented a case where such a stocking is developed using a blend yarn consists of selected shape-memory polyurethane and nylon filaments. Extensive experimental work has also been conducted to demonstrate the feasibility and explore the influencing factors involved.

**Keywords:** Compression stocking; compression therapy; shape memory polymer; venous ulcers; interface pressure; heat stimuli; rubber elasticity.

## 1. Introduction

Patients suffering from chronic venous disorders, such as leg ulcers, oedema, venous stasis, venous hypertension, etc., are known to have poor quality of life due to continuous discomfort or pain, limited mobility and long recovery time, in addition to a rigorous management plan and thus the financial cost involved [1, 2]. Compression therapy is the cornerstone in the conservative treatment of such disorders since ancient times. Herein, external compression is provided to the affected leg by applying medical stockings or bandages to accelerate venous blood circulation, and finally decrease the venous pressure. The success of this treatment depends to a great degree on the level of pressure at the affected portion on the limb, and the sustenance of this pressure during the course of treatment [3, 4]. This interface pressure has to be applied quite accurately within certain limits and should not be either below or above the prescribed level, or certain complications will occur [5, 6].

In practice, selection of the stockings with proper sizing and fitting has always been a contest for both health practitioners and manufactures, for better patient's compliance and more effective treatment [7]. Different class of stockings are required to provide light (Class I, 14-17 mmHg), medium (Class II, 18-24 mmHg) and strong (Class III, 25-35 mmHg) levels of compression depending on the severity of the disease [8]. However in practice it is difficult to achieve the targeted pressure level due to various reasons, including mainly the different leg attributes (shape or size) among patients, and difference in material (including both stockings and the legs) properties (time and temperature dependence). Moreover, pressure drop over time is also a major concern due to the time dependence of the system behaviours [9]. For instance, experimental studies have showed that the pressure decreases over time due to reduction in swelling [10]. Also, many compression products displays initial pressure drop just after their application [4, 11], as they are made of polymeric materials (cotton, viscose, PET, etc.), to which stress relaxation is an inherent attribute, although stockings containing elastomeric yarns can alleviate the problem. As pressure drop is inevitable for almost all available stockings, and re-instalment or replacement of the stocking is needed once the pressure falls below the targeted level. Another reason for changing the stocking is for patient comfort, as there is often the need during night the stocking be removed so it will not interfere with the sleep [12]. Clearly, the aforesaid inadequacies of the conventional approaches demonstrate a compelling demand for a novel smart stocking system in compression management that allows modulating the compression level via external control to easily change or readjust pressure whenever needed.

In this work, based on some theoretical considerations, we first derived several criteria that have to be satisfied simultaneously in order to achieve the controlled pressure adjustment using external heat stimuli. We then proposed the use of shape memory polymers for the development of a smart compression stocking. Shape memory polymers (SMPs) are the smart material that can memorize the original shape so that they can recover from a temporary deformed shape, upon exposure to an external stimulus, e.g., heat, light, water, etc. [13]. SMPs have gained practical significance over the last 10 years in developing many potentially innovative products for biomedical applications such as clot removal devices, aneurysm occlusion devices, vascular stents, orthodontics, tissue engineering, etc. [14-28]. In the field of compression management, shape-memory polymer based film actuator has been proposed by Ahmad et al. (2012) for pressure bandage application. They have used temperature-responsive SMP strips attached to fabrics to control the compression by an external heat source. The use of SMP film actuators may significantly obstruct the moisture transmission and the permeability of the compression system, and therefore not a viable solution for providing improved comfort to the patients. Moreover, the use of shape memory alloy based compression bandage as proposed by Moein and Menon (2014) is also not an effective method as there exists challenges in the integration of shape memory alloy wires with the textile yarns.

A thermal sensitive SMP has the potential to adjust the internal stress in its structure via external heating [29-31] and this characteristic is the key for the present case to obtain smart compression management as suggested by Laplace's law [32-34]. We will investigate in this paper the fundamental relationship between the extra pressure and the recovery stress generated by the stocking. We will then present a case where such a stocking is actually developed using a blend yarn consists of selected shape-memory polyurethane and nylon filaments. Extensive experimental work has also been conducted to demonstrate the feasibility and explore the influencing factors involved.

## **2. Working principle and material determination**

Before proceeding with the stocking design, there are a couple of theoretical issues to be examined to provide guidance in selection of desirable material.

### *2.1. Leg, stocking and generated pressure*

The pressure developed in the leg by wearing a stocking depends on the shape, size and the particular position of the leg. Once the stocking is applied to the leg, a tensile strain in the stocking,  $\varepsilon$ , is generated and can be calculated as,

$$\varepsilon = \frac{C_l - C_s}{C_s} \quad (1)$$

Where  $C_l$  and  $C_s$  are the original circumferences of the leg and the stocking, respectively, and  $C_l > C_s$ . In other words, for a given leg size  $C_l$ , this strain value is determined, exclusively, by the stocking size  $C_s$ , if we ignore the minimal effect of the stocking thickness. The corresponding stress  $\sigma$  in the stocking can be obtained simply as

$$\sigma = E \times \varepsilon \quad (2)$$

where  $E$  is the tensile modulus of the stocking. Combining Eqs. 1 and 2 and according to Laplace's law [34], an internal radial pressure  $P$  will be exerted by the tensioned stocking on to the leg as

$$P = \frac{\sigma w}{r} = \frac{E \varepsilon w}{r} = 2\pi E w \left( \frac{C_l - C_s}{C_l C_s} \right) \quad (3)$$

where  $r = \frac{C_l}{2\pi}$  is the radius of the leg and  $w$  is thickness of the stocking. So our problem of applying a proper level of pressure  $P$  to the leg of size  $C_l$  via a stocking appears to be a simple matter of selecting a stocking size  $C_s$  based on Eq. 3. However there are several factors that complicate the process:

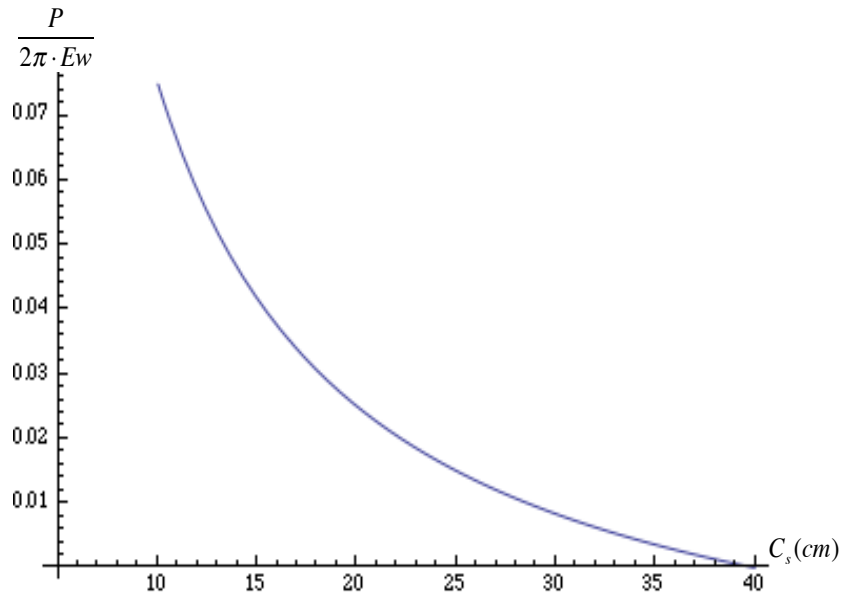
- Most importantly, the stocking materials are viscoelastic **whose properties are time and temperature sensitive**. Similar concern exists to the material nature of the human body that is made of mostly biopolymers as well. This means that due to the stress relaxation or strain creep in such materials, both strain and stress developed in the system are going to fade away with time. So even we selected a stocking size  $C_s$  to achieve the desired pressure level  $P$  to the leg using Eq. 3 initially, the initial tensile strain  $\varepsilon$  developed will diminish gradually with time because of the creep, leading to the decline of the pressure level.
- The tensile modulus  $E$  of the stocking and its equivalent counterpart of the leg are both **heat sensitive** and will also alter with the interfacial temperature between the leg and the ambient.
- To make the matter worse, our leg is not a single sized solid cylinder but with different circumferences at different locations, and often **different pressure levels**

are needed at different locations of the leg. A gradient pressure with high pressure at ankle and low at knee is frequently required [35]. This demands a proper size fitting or customized stocking to a particular leg.

- **Varying shape or size of legs for different patients** increases the complexities at manufacturing level as different choices for stocking size should be available to the clinicians for different patients. This further adds confusion to nurses in the selection process and also increases the cost to the manufacturers.
- In addition, **different compression is required at different stage of venous disease**. For example, 14-17 mmHg is required at the stage of varicose veins while 25-35 mmHg is usually recommended at the stage of venous ulcers. This means we need different stocking sizes even for the same leg depending on the pressure requirement.
- A lesser but worthy point is about the nonlinear influence of change stocking size  $C_s$  on the resulted pressure level  $P$  for a fixed leg size  $C_l$ . If we rearrange Eq. 3 into

$$\frac{P}{2\pi Ew} = \left( \frac{C_l - C_s}{C_l C_s} \right) \quad (3b)$$

and plot it in Fig. 1 where leg size  $C_l = 40$  cm.



**Fig. 1:** Nonlinear relation between pressure and stocking circumference.

That is, even at a fixed temperature level so the tensile modulus  $E$  remains constant, **the relationship between the pressure  $P$  and stocking size  $C_s$  is not linear**, revealing why it takes some training and practice for a nurse to get even the initial pressure right.

To conclude, giving the complexities discussed above, it is difficult if possible to achieve and maintain a desirable pressure level in treating chronic venous disease using the existing stockings. If looking at Eq. 3 more closely however, the key factor causing the diminishing pressure  $P$  is the relaxed stress  $\sigma$  in the stocking. If we can find a material whose internal stress  $\sigma$  can be easily adjusted in such that it can compensate the deviations of the pressure from the initially designated level, we can then sustain the desired pressure on the leg. Furthermore, we can even modulate the internal stress  $\sigma$  so as to eliminate the need of multiple stocking sizes in achieving targeted pressures, i.e., a smart stocking!

## 2.2. The desired temperature, modulus and pressure relationships

In the quest for such materials with adjustable internal stress  $\sigma$ , polymeric materials are the logical choice for it is widely known that their tensile modulus  $E$ , i.e., internal stress  $\sigma$ , can be altered if we control the temperature during operation. However not all polymers can achieve this smart stocking function: after all, cotton and nylon used in current stockings are already polymers themselves. **We need polymers with internal stress  $\sigma$  exceptionally sensitive to temperature change within a very narrow range around body temperature.**

To analyse the connection between temperature and internal stress, assume we have selected a material to which the rubber elasticity theory is valid. Since the deformation of rubber is largely governed by entropy elasticity, the contribution of energy part can be ignored. This will result in the following relation between change in material stress  $\sigma$  caused by temperature  $T$ ,  $\frac{\sigma}{T}$ , in reference [36]:

$$\left(\frac{\partial \sigma}{\partial T}\right)_{l,V} = \frac{\sigma}{T}, \quad (4)$$

if we keep the sample length  $l$  and volume  $V$  constant. The general solution for Eq. 4 can be stated as:

$$\sigma = a + bT \quad (5)$$

Where  $a$ , and  $b$  are both constants. It is clear from the result that how temperature  $T$  change affects the internal tension  $\sigma$  depends on the two constants  $a$ , and  $b$ . Experimental data has showed [36] that by applying a pre-strain  $\varepsilon$  on the rubber, we can adjust the response of the stress toward temperature change, i.e.:

1. If the pre-strain is at a critical level  $\varepsilon = \varepsilon_c$ , Eq. 5 will reduce to

$$\sigma = \sigma_o \quad (5a)$$

That is  $a = b = 0$ , so that the internal tension  $\sigma$  will remain constant  $\sigma_o$ , independent of the temperature change;

2. If however there is  $\varepsilon < \varepsilon_o$ , the equation can be expressed as

$$\sigma = a - cT \quad (5b)$$

where  $a > 0$  and  $b = -c > 0$  but still a constant. The stress  $\sigma$  will decrease with the temperature;

3. If there is  $\varepsilon > \varepsilon_o$ , we will have

$$\sigma = a + bT \quad (5c)$$

So the stress  $\sigma$  will increase with the temperature. This is the behavior we are seeking - by raising the temperature, we can increase the internal stress  $\sigma$ . **So to assure such a material will act as designated in Eq. 5c, a pre-applied strain  $\varepsilon > \varepsilon_o$  is required, where  $\varepsilon_o$  can be easily determined through experiment.**

Then once such a stocking is produced, and put on the leg by applying a pre-strain  $\varepsilon > \varepsilon_o$ , a corresponding stress  $\sigma$  and hence compression  $P$  will be applied to the leg: the system is in equilibrium. If a stress dropping  $\Delta\sigma$  occurs and to compensate for that, we need a temperature increase  $\Delta T$  according to

$$\Delta\sigma = b\Delta T \quad (6)$$

where  $b > 0$ . The corresponding increase in the pressure is

$$\Delta P = \frac{w\Delta\sigma}{r} = \frac{wb\Delta T}{r} \quad (7)$$

For given  $r$ ,  $b$  and  $w$  there is a linear relation between  $\Delta P$  and  $\Delta T$ . Eqs. 5c and 7 can thus be used as the criteria in material selection for the smart stocking.

### 2.3. The potential materials

To sum up, we need a polymer material:

1. whose mechanical properties are sensitive enough to temperature changes of a narrow range above the human body temperature;



2. It obeys the rubber elasticity theories and, more specifically, follows Eqs. 5c or 6: during use of the stocking, an increase in temperature  $\Delta T$  will lead to a sufficient increase in internal stress  $\Delta\sigma$  - hence  $a, b > 0$ , and parameter  $b$  is sufficiently large;
3. Also the selected material should be easy to process into stockings and comfort to wear in contact with skin.

So although natural rubber satisfies the theories but it requires temperature change much higher than body temperature to generate sufficient compensating stress. Also it requires relatively large deformation to be used. In addition, rubbers are not as easily processed into fibers and not comfortable to wear. Then shape memory polymers (SMP), or more specifically the shape memory polyurethane (SMPU), came to our attention. First, the deformation of SMPU is largely governed by entropy elasticity [37] and thus the rubber theories are expected to be valid to it. SMPU are stimuli-responsive materials and have demonstrated sufficient changes in mechanical properties over desired temperature range. Finally they are easy to process into comfortable stockings.

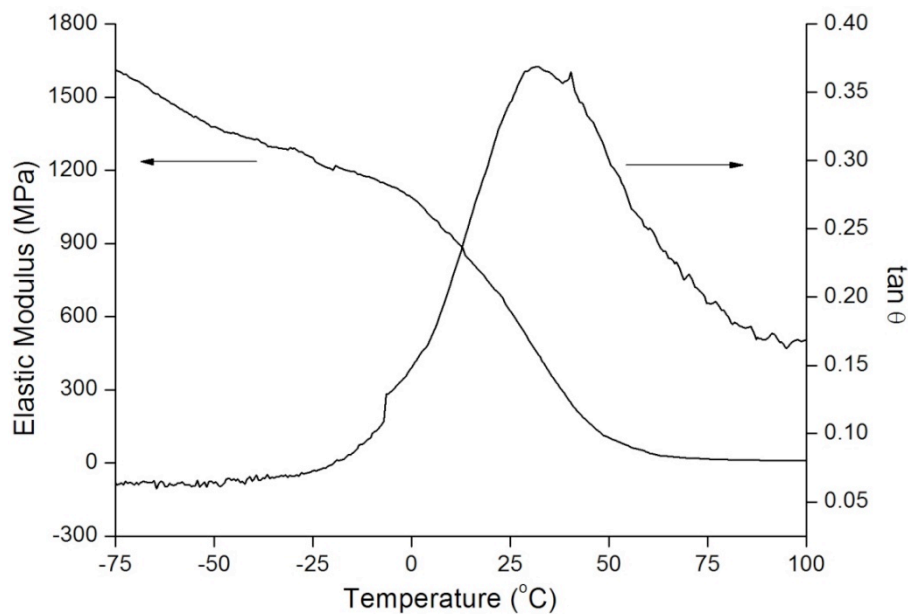
### **3. Materials and experimental methods**

#### *3.1. Preparation of SMP chips*

The shape memory polyurethane (SMPU) chips was prepared by bulk polymerization method using polytetramethylene ether glycol (PTMEG;  $M_n = 650$ ; Aldrich Chemical Company, USA) as the soft segment, and 4,4'-methylene diphenyl diisocyanate (MDI; Aldrich Chemical Company, USA) and 1,4-butanediol (BDO; Acros Organics) as hard segment. The weight ratio of soft segment and hard segment was 12:13. Extra pure-grade MDI was used for the synthesis without further treatment. BDO (1,4-butanediol) was dried by 4 Å molecular sieves beforehand. PTMEG were dried and degassed at 80 °C under 0.1–0.2 kPa for 12 h prior to be used. Initially PTMEG and MDI were kept for reaction at 80 °C for a period of 2 hrs. After the formation of pre-polymer, BDO was added as a chain extender and kept for 1 min to complete the reaction. A nitrogen environment was maintained for all the reactions. The reaction temperature was controlled to be lower than 90 °C. The reacted polymer was poured into a pre-heated (100 °C) polytetrafluoroethylene mould and a PU sheet (~3.0 mm-thick) was obtained. The sheet was then incubated in a vacuum oven for 24 h at 100 °C. SMPU chips was then obtained by chopping the sheet and extruded using a single-screw extruder followed by pelletizer.

#### *3.2. Development of SMPU filament and smart stocking*

Melt spinning process was employed to spin the filaments from the prepared SMPU chips. Prior to spinning, the SMPU chips were cured for 24 hr at 100 °C until the moisture level reached less than 100 ppm (parts per million). The SMPU filaments were spun in highly pure nitrogen environment using single screw extruder with a temperature range from 175 to 202 °C. The winding speed was set at 500 m/min, and the obtained linear density of the filaments was 18.6 tex. A dynamic mechanical analysis test was performed using a Perkin–Elmer diamond dynamic mechanical analyser operated in the tensile mode to find the thermal transition ranges for the filament (Meng et al., 2007). The variation of elastic modulus ( $E'$ ) and loss tangent ( $\tan \theta$ ) over the temperature range from  $-75$  to  $100$  °C are showed in Fig. 2. The  $T_g$  of the SMPU is thus around  $30$  °C, the activation temperature close to the body surface temperature.



**Fig. 2** – DMA results of the SMPU filament.

A combined yarn consisted of SMPU (18.6 tex) and Nylon (18.9 tex) filaments was used for making a smart stocking using a circular knitting machine. The structural details of the knitted stocking are listed in Table 1. The machine gauge used was 21 needles per inch.

**Table 1.** Details of the stocking.

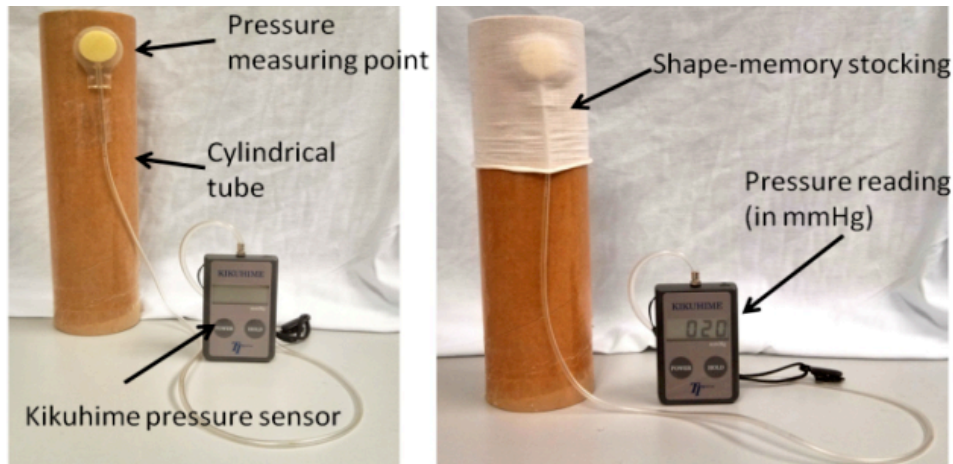
<b>Weave type</b>	Circular knitted
<b>Linear density of yarn used; tex</b>	18.9 (Nylon) 18.6 (SMPU filament)
<b>Chemical composition; %</b>	50.4 (Nylon) 49.6 (SMPU filament)
<b>Mass per unit area; g/m<sup>2</sup></b>	310.1
<b>Thickness; mm</b>	0.57
<b>Threads per unit length</b>	12 (wales/cm) 27 (courses/cm)
<b>Circumference; cm</b>	22.2

**Note:** tex is the weight (in gm) for 1000 m of the yarn.

Prior to using the stocking as a credible compression product, we have to examine its related performance, and the subsequent sections describe the characterization of the stocking properties.

### 3.3. *Interfacial Pressure measurement*

The interfacial pressure caused by the stocking was measured using a Kikuhime<sup>TM</sup> pressure sensor, and the experimental set-up is shown in Fig. 3. Cylindrical tubes of different circumferences were used to act as a human leg. Before the application, the stocking was heated for 10 min using a hair drier and then applied over the cylindrical tube. The system was then allowed to cool down at room temperature for 1 hr. A baseline pressure as in Eq. 3b was obtained at the room temperature, owing to the elastic stress caused by the stretch in the filaments in the fabric structure, although the internal stress and hence the generated pressure started to decay immediately. To compensate the stress relaxation, the entire set-up was placed in a heated chamber for the heat stimulus  $\Delta T$ , which causes an increment stress  $\Delta\sigma$  and hence the compensating pressure  $\Delta P$ . The net increase in the interface pressure  $P$  above the baseline pressure was then recorded at three levels each for the two factors, temperature (30, 40 and 50 °C) and pre-strain  $\varepsilon$  (5.85, 13.06 and 20.27 %). The pressure variation over time was also examined to evaluate its longer term performance.



**Fig. 3** – Set-up for pressure measurement

#### 3.4. Measurement of the recovery stress $\Delta\sigma$

To explain the increase in pressure of the stocking, we have to verify the idea of increasing recovery stress  $\Delta\sigma$  at elevated temperature  $\Delta T$ . Even though Eq. 6 offer the connection between the two, we still employed a thermo-mechanical uniaxial tensile testing to validate the occurrence of  $\Delta\sigma > 0$  at a given  $\Delta T$ . The measurement was done using a tensile tester (Instron 5566), for loading and unloading, anchored with a temperature chamber for heating and cooling. A size of 150 mm length and 50 mm in width was cut along the circumference of the stocking, and used for testing. The specimen gauge length was 100 mm. The tensile load was measured using a load cell and the displacement of the gauge length was measured from the displacement of a cross-head. Initially, the clammed specimen was first heated above  $50\text{ }^{\circ}\text{C}$  ( $> T_g$ ) in the chamber and then stretched to a particular extension level in the heated condition. It was then allowed to cool down at room temperature for 1 hr in the extended state. Thereafter, the specimen was heated again under the constant extension for the activation of the recovery stress and the variation in the stress was recorded at the similar levels of temperature (30, 40 and  $50\text{ }^{\circ}\text{C}$ ) and pre-strain (6, 13 and 20 %) used for stocking pressure  $P$  measurement. ANOVA (analysis of variance) analysis was performed for the test of significance, and a factor was considered as statistically significant if its p-value is less than 0.05. The linear regression analysis was done to find the correlation between the maximum value of extra pressure generated by the stocking and the results for the maximum recovery stress obtained at different temperature and strain levels.

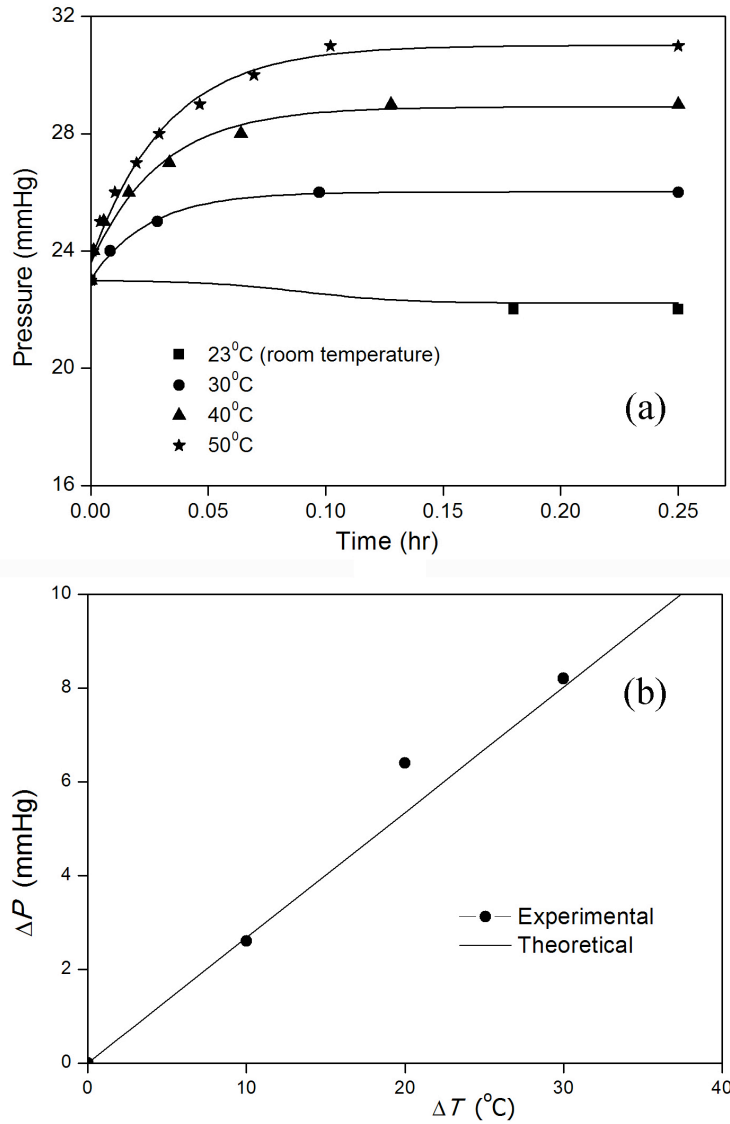
## 4. Results and discussion

Clearly, giving the nature of both the SMPU itself and the stocking-leg system, and when the patient leg and stocking design and size are fixed, the pressure level actually exerted to the leg by the stockings is determined by the following major factors.

#### 4.1. *Effect of the activation temperature*

As the stocking is in contact with human body, the design for the desired range of  $T_g$  is important. By proper choice of different components (type and molecular weight of switching segment; hard segment content) involved in the formation of the SMP, the  $T_g$  can be easily adjusted to a particular targeted range [38, 39]. Herein, we developed shape-memory polyurethane (SMPU) filament with low  $T_g$  ( $\sim 30^\circ\text{C}$ ). The range for the activation was chosen from  $30^\circ\text{C}$  up to  $50^\circ\text{C}$ .

The pressure  $P$  at different level of temperature was showed in Fig. 4. For a particular temperature, the pressure in Fig. 4a increases for the initial period (10 min) and finally reaches to a fixed value. The rate and amount of extra pressure generated is more at larger temperature ( $p < 0.01$ ). SMPU filaments had shown a range of transition temperature (Fig. 3) due to the fact that not all the switching segments would activate altogether at a fixed temperature point. It is expected that more switching segments are activated at upper extreme of  $T_{\text{trans}}$  ( $\sim 50^\circ\text{C}$ ) as compared to its lower extreme ( $\sim 30^\circ\text{C}$ ). Therefore, more amount of recovery stress will be generated in the SMPU filament at higher temperature. Fig. 4a also reveals that the slope of the pressure-time curve (the modulus  $E$ ) of the stocking is indeed increasing with  $T$  as desired. Furthermore the plot in Fig. 4b shows a linear relationship between  $\Delta P$  and  $\Delta T$  as dictated by Eq. 7. **Fig. 4 thus confirms first, our SMPU obeys the rubber elasticity theories, and more importantly we can indeed adjust the stocking stress by external temperature stimuli!**



**Fig. 4** – (a) Pressure variation at different levels of temperature (13% strain). (b) Comparison with theoretical prediction using Eq. (7).

#### 4.2. Effect of initial strain

A minimum strain  $\varepsilon_0$  has to be imposed in the new stocking to obtain increased stress or pressure with increasing temperature. The choice of initial strain  $\varepsilon_0$  is hence critical for the pressure control and its value is determined by two competing factors in a stretched SMPU specimen at elevated temperature. First this specimen, like other objects, will expand once heated. On the other hand, the internal stress in a stretched SMPU under constraint is caused by the entropic elasticity that will facilitate thermal contraction at high temperature. So both thermal expansion and contraction take place simultaneously, depending on the level of strain imposed. At low strain, thermal expansion will be predominant, but thermal contraction will increase at higher strain level.  $\varepsilon_0$  thus represents the strain level at which the

expansion and contraction cancel each other and the internal stress becomes independent of temperature. The thermal strain of different SMPUs were reported in the range from 1 to 3% in our earlier study [40]. For the present SMPU, the thermal strain was found to be around 2.5% in the temperature range from 20 to 50°C. Also a minimum of 15 mmHg initial pressure is required for a Class I stocking, we hence selected a minimum 5% strain in the stocking to achieve desirable results.

The effect of strain levels in the stocking on the pressure variation is shown in Fig. 5. Three different levels of strain were obtained in the stocking by wrapping it on three different cylindrical tubes having different circumferences (23.5, 25.1 and 26.7cm). More circumferential strain was resulted when the stocking was wrapped on the cylinder of higher circumference, and this provided more initial compression, i.e., baseline pressure. It is found that more amount of extra pressure above the baseline pressure level is generated with increasing strain in the stocking ( $p < 0.01$ ). **Clearly, all three strain levels are sufficient for the SMPU to act following Eq. 5c, and a higher strain level leads to a greater parameter  $b$  and hence a higher  $\Delta P$ . The  $b$  values obtained using Eq. (6) for 6, 13 and 20% are 1.95, 2.5 and 4.59 kPa/°C respectively.**

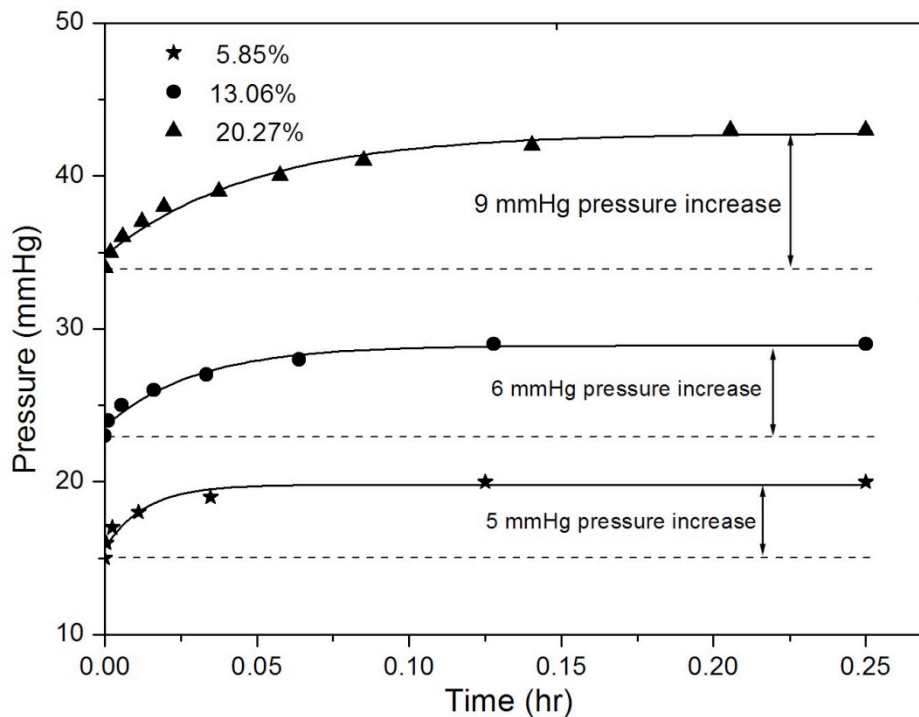


Fig. 5 – Pressure variation at different levels of strains ( $T = 40^\circ\text{C}$ ).

#### 4.3. Correlation of extra pressure and recovery stress

As suggested by Laplace's law (or Eq. 7), the interface pressure depends on the internal tension or stress developed in the stocking in the wrapped position. The change in the internal stress  $\Delta\sigma$  in the stocking will result in pressure variation  $\Delta P$ . To elucidate these facts, we performed the correlation tests of the variation of pressure and tensile recovery stress over time during the activation of stocking. A high value of coefficient of determination (0.96) was found between the data of maximum recovery stress and extra pressure generated at different temperatures and strain levels. Table 2 shows the result of maximum recovery stress upon activation of the stocking. This clearly indicates the potential of the stocking to vary the pressure to a wide range. Table 3 shows that the stocking is able to generate extra pressure up to maximum of 16.2 mmHg by selecting suitable level of temperature and strain. That is, for a base line pressure of 30 mmHg, we can achieve maximum pressure up to 46.2 mmHg, more than 50% increase.

**Table 2.** Maximum recovery stress  $\Delta\sigma$  ( $10^4$  Pa) generated by the stocking upon activation at different levels of temperature and strain

Pre-strain	Temperature ( $^{\circ}\text{C}$ )		
	30	40	50
6%	1.71 (0.12)	4.39 (0.35)	5.61(0.33)
13%	2.63 (0.22)	5.32 (0.31)	7.24 (0.6)
20%	5.87 (0.44)	9.42 (0.64)	13.22 (0.75)

**Note:** Values in the bracket represent coefficient of variation (in %)

**Table 3.** Maximum level of extra pressure  $\Delta P$  (in mmHg) generated by the stocking upon activation at different levels of temperature and strain

Strain	Temperature ( $^{\circ}\text{C}$ )		
	30	40	50
5.85%	1.8 (0.13)	5.2 (0.41)	6.8 (0.47)
13.06%	2.6 (0.31)	6.4 (0.44)	8.2 (0.54)
20.27%	5.4 (0.39)	9.2 (0.58)	16.2 (0.84)

**Note:** Values in the bracket represent coefficient of variation (in %)

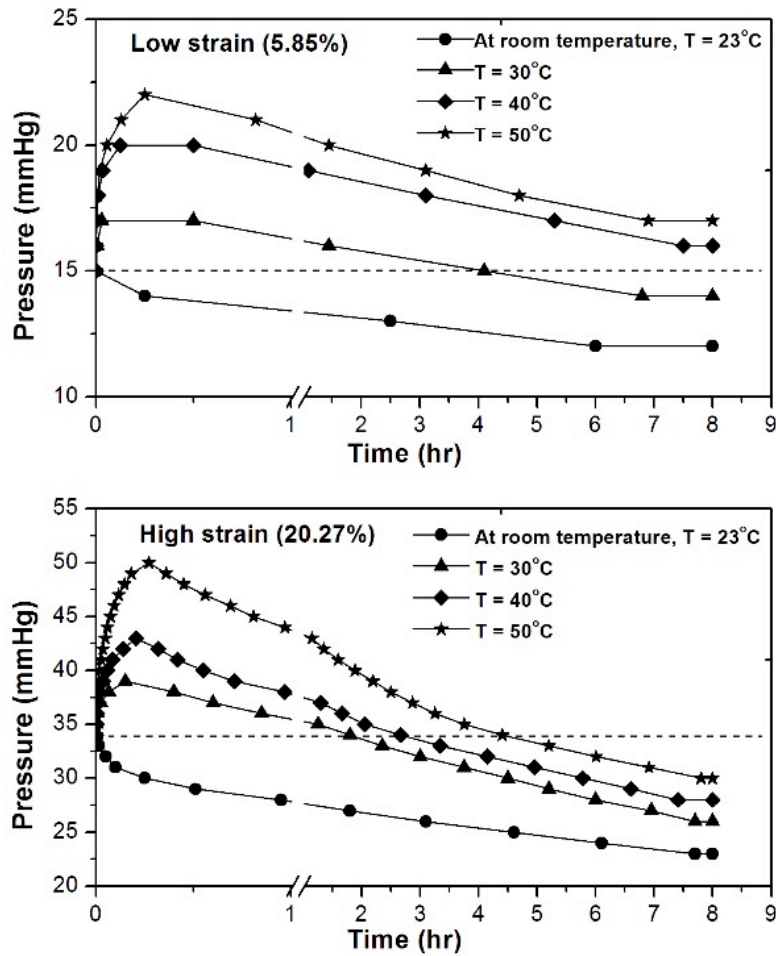
#### 4.4. Time dependence property

Continuous compression should be maintained for optimum health benefits, and stocking should sustain the pressure in recommended range for an extended period of time.



Herein, we also investigated the longer term compression performance of the stocking. Fig. 6 shows the pressure variation over 8 hrs at different levels of temperature. At room temperature, a pressure drop of 3 mmHg is observed in 8 hr. Pressure drop is faster for initial period, and then slows down. On activating the stocking at greater temperature, the pressure increases to a higher level for the initial period (10 min), and thereafter starts decreasing. The nature of pressure drop at higher temperature is also similar to that of observed at room temperature. Examining pressure variation at different strains, it was found that the pressure drop was more at high strain level (32% pressure drop at 20.27%) compared to low strain (20% pressure drop at 5.85%) at room temperature. The pressure reduction in the stocking is mainly due to the relaxation of internal stress in the nylon and SMPU filaments over time [11, 32]. Also, the relaxation in the fiber increases at higher strain level [4]. Although an inherent feature of all polymers, the magnitude of relaxation is more for non-elastomeric polymers (cotton, viscose, PET, nylon, etc.; > 30% relaxation), as compared to elastomeric samples (polyurethane; < 10% relaxation) [4, 41]. Given this fact, the relaxation presented in this work could be mainly caused by Nylon (with around 50% of the fiber contents in the stocking fabric) and increasing the SMPU filaments will alleviate the problem as discussed so far.

To further minimize the relaxation, an in-lay structure could be created for the stocking where the SMPU acts as a load bearing yarn and the nylon as loop forming yarn with less stretch and hence minor relaxation. Additionally, we have proposed a thermomechanical method to remove the relaxation in the SMPU and achieve pure elastic recovery stress [42].

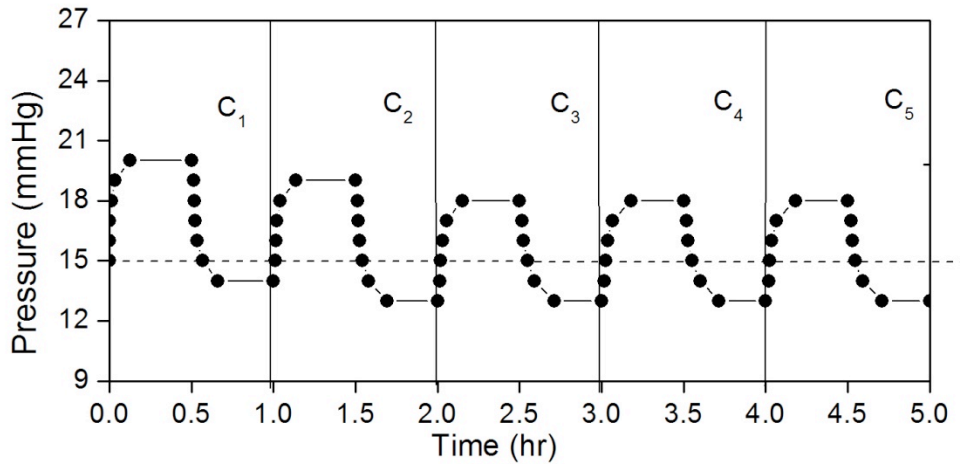


**Fig. 6** – Pressure variation over time at different levels of strain and temperature.  
 (a) Top - low strain level 5.85%, (b) Bottom - high strain level 20.27%.

#### 4.5. Cyclic tests

Finally, we demonstrated the ability of the stocking in reproducing or readjusting the pressure when used multiple times. For the cyclic tests, the stocking was activated for 30 min in heated chamber, and then placed at room temperature for 30 min. The procedure was repeated for 5 times, and the pressure variation was recorded. Repeatability tests were also performed at different strain levels and at different chamber temperature levels used during heating of each cycle. The nature of pressure variation over 5 cycles is summarized in Fig. 7. There is initial readjustment of pressure between the first two cycles. In the first cycle, the amount of pressure increase during heating is less than the pressure decrease during cooling. This resulted in lowering of base line pressure (initial pressure at room temperature) at the end of the first cycle. This effect continued up to first two cycles and the pressure variations

become alike for subsequent cycles. Similar results were obtained at different strain levels. However, the decrease of baseline pressures was more at high strain level compared to low strain. The pressure drops of 10 and 20% were obtained respectively at 5.87 and 20.27% strain levels. No significant variation was obtained from third cycle onwards. The stocking showed the potential of reproducing the same amount of pressure even after multiple uses.



**Fig. 7** – Pressure variation during sequential heating and cooling of the stocking up to 5 cycles (C<sub>1</sub>-C<sub>5</sub>: individual cycles). Each cycle includes heating of the stocking for 30 min in heated chamber (T = 40°C), and then cooling down for another 30 min at room temperature (T = 23°C).

#### 4.6. Discussions and remaining challenges

The methodology in determining SMPU and stocking size is proposed in the study, and it offers several potential advantages in medical compression management:

1. Based on Eq. 6, the internal stress in the SMPU stockings can be easily increased by heat stimuli. So when the stocking pressure drops below a targeted level, it can be readjusted via a heat stimulus without replacing the stocking, thus provide sustained compression for longer period;
2. In the current practice, even for the same patient, different range of stockings are required based on limb shape or size, and level of compression. The temperature-pressure re-adjustment of our new stockings will definitely reduce the stocking ranges needed to achieve the required pressure levels;
3. Also, the temperature-pressure sensitivity of our new stockings will facilitate nurses for easy application of stocking to limb. Here, we have the advantage for easy application of the stocking providing low compression (Class II or I) initially, and later high compression (Class III) could be achieved by simple heating;

4. The new stocking could provide better compliance for patients owing to the easily re-adjustable pressure;
5. Because of the internal stress and heat stimuli connection of SMPU materials, stockings with massage effect can be easily developed if a programmed heating source is incorporated into the system.

Naturally at this initial stage, there remain a few technical challenges before to ensure the stocking as a credible compression product. The  $T_g$  of the SMPU filament needs to be controlled further to allow easy activation at still lower temperature range. Also in real practice, heating a stocking on the leg to a particular temperature is a challenge, and simple and efficient heating arrangement need to be assured to have precise control on pressure. Note that although pressure drop in the smart stocking can be compensated under heat stimuli, it can also be minimized by increasing the proportion of elastomeric SMPU fibers in the structure. However, increasing the elastomeric fibers makes the structure tighter and less permeable, and hence diminishes the comfort, and thus a proper blending with cotton or other fibers is preferred for comfort. Optimization should be done to obtain both desirable compression and sufficient comfort. Future work in SMP should be done to examine several other factors, e.g., different SMPs, hard segment content, etc.

## 5. Conclusions

First in smart stocking material selection, the following criteria have to be met simultaneously, based on the methodology developed in this paper:

1. Its mechanical properties are sensitive enough to temperature changes of a narrow range above the human body temperature;
2. It obeys the rubber elasticity theories and, more specifically, follows Eqs. 6: during use of the stocking, an increase in temperature  $\Delta T$  will lead to a sufficient increase in internal stress  $\Delta\sigma$ ;
3. Also the selected material should be easy to process into stockings and comfort to wear in contact with skin.

The above criteria render the shape memory polymers superior to other similar materials including rubber. A smart compression stocking using SMPU has been successfully developed and examined for the pressure management. It was developed using a blend yarn consists of shape-memory polyurethane and nylon filaments so that the internal stress in its structure can be changed by external heat. Our experimental results have demonstrated that

this product indeed allows controlling or managing the pressure exerted by the stocking in wrapped position, and extra pressure (up to 50%) can be generated by simply heating the stocking. Such a stocking has the potential to overcome the limitation of conventional stockings as it could provide the freedom to adjust pressure level externally whenever needed, i.e., a smart wound care product, during the course of compression therapy.

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