3D FORMATION OF BIOINSPIRED TEMPERATURE-RESPONSIVE MATERIALS AND THE FABRICATION PROCESS THEREOF

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ABSTRACT

The responsive behaviour of non-living tissues under an external stimulus is determined by the anisotropic, non-homogenous and multilayer characteristics of their structure. The development of responsive materials that alter their shapes in response to environmental stimuli is of great importance. Most of the proposed materials or structures are fabricated as 2D plane surfaces and alter their geometry to become 3D complex structures in response to a stimulus. In this study, we present the fabrication process and the transformation mechanisms of multilayer materials that have been formed to have a 3D initial shape and alter their geometry to form 2D shapes or other complex 3D shapes without the use of complex forming tools. The shape change is reversible under a temperature stimulus. This approach uses the multilayer structure, the anisotropic thermo-mechanical properties of the different materials, and the variable modulus of elasticity of the polymeric materials in order to create responsive 3D complex geometries at room temperature. Finally, we demonstrate a complex structure that alters its shape from a 3D shape to a 2D or to a different 3D shape and we study the curvature and the occurred failures during the fabrication process.

Keywords: Responsive materials, Smart materials, Bioinspired materials, Anisotropy

1. INTRODUCTION

Non-living tissues with very complex geometrical characteristics transform their shape owing to the coefficients of hygroscopic and thermal expansion of their multilayer and anisotropic structure [1–4]. The transformation of the aforementioned non-living tissues can be mimicked and controlled through the use of multi-layered anisotropic materials, nano-reinforced multilayer hydrogels, pre-stressed sheets, fibrous composites, and anisotropically patterned multilayer materials [5–15]. The development of the responsive materials that alter their shapes in response to environmental stimuli is of great importance. Most of the proposed materials or structures are fabricated as 2D plane surfaces and alter their geometry to become 3D complex structures. In this case, a material with a 2D geometry alters its shape to assume a 3D complex geometry in response to a stimulus [5–15]. The geometry of these materials can be transformed under a temperature stimulus or humidity. The present authors have developed low-cost anisotropic multilayer films that largely deform when a temperature change occurs; this behaviour is attributed to the mismatch in the coefficient of thermal expansion between the film layers and to the anisotropic structure of the layers that causes the transformation of the material [13-15] in order to control the temperature of a body. In all aforementioned studies and strategies, the materials transform from a 2D shape (sheet) to a 3D shape.

In this study, we will present two different fabrication processes for the development of 3D complex responsive materials, where the 3D geometry was formed at room temperature (initial shape) and then transformed to a 2D geometry as the temperature increased. The proposed responsive multilayer anisotropic materials were able to passively react under temperature stimuli by transforming their geometry from 3D shapes to 2D shapes. This approach bypasses the use of complex tools to form the geometry of the structure in 3D space. The 3D-shape configuration was programmed by using the multilayer structure and its anisotropic thermo-mechanical properties (anisotropic coefficient of thermal expansion and anisotropic modulus of elasticity) of the different materials and the variable modulus of elasticity of the polymeric materials in order to create responsive 3D complex geometries at room temperature. The anisotropy of the layers controls the shaping modes of the material during its transformation. We will present the different transformation steps of the materials, as well as their changes in their curvature during this transformation. A preliminary scanning electron microscopy (SEM) characterisation revealed the crack formation, which was caused on the anisotropic metallic strips. Finally, to demonstrate the capabilities of the proposed materials, we will present a more complex structure (flower), whose initial shape is a 3D geometry at room temperature and which then transformed to a different complex shape and returns to its initial position.

2. FABRICATION PROCESSES AND SHAPE TRANSFORMATION

The multilayer film consists of a high-CTE anisotropic layer (oriented polyethylene), one layer of polymeric adhesive, and aluminium strips laid in a certain direction (low-CTE material). The anisotropy of the layers and the mismatch in the coefficient of thermal expansion between the layers are the main parameters for the control of the geometry of the material as the temperature increases. Fig. 1b illustrates in detail the sequence of the anisotropic layers and the strips of the passive and responsive / active regions that were used. The multilayer material consists of aluminium strips (thickness ≈ 5 μm) over a bilayer material (41 μm polyethylene and adhesive). The deformable regions are very responsive to temperature and present extremely large deformations, Fig. 1a. Also, other thicknesses can be used.

Two major fabrication steps are required in order to form 3D responsive layers at room temperature. The first step was to form the aluminium strips on the polymeric substrate. The materials were manufactured on a 2D plane and transformed to 3D shapes under temperature stimuli, Fig. 1a. The direction of the strips, the direction of the oriented polyethylene (PE), and the thermo-mechanical properties of the material determine how the shape will be transformed in 3D space. The aforementioned parameters control only the transformation of the geometry. The second step was to take advantage of the variable and temperature-dependent modulus of elasticity of the polymeric material. After a critical temperature level is exceeded, the 3D geometry of the multilayer material transforms to a 2D geometry. During the cooling stage, the multilayer material transforms back to a 2D geometry until it reaches a steady state temperature. Thus, at room temperature, the shape of the multilayer material has been transformed to a 3D shape (first figure of Fig. 1b). The material remained functional and transformed from a 3D shape to a 2D shape or a different 3D shape, Fig. 1b. It should be mentioned that the thermo-mechanical properties had changed before and after the critical temperature level had been reached. The direction of the strips, the oriented PE, and the altered thermo-mechanical properties of the material determine how the shape will be transformed in 3D space at room temperature.

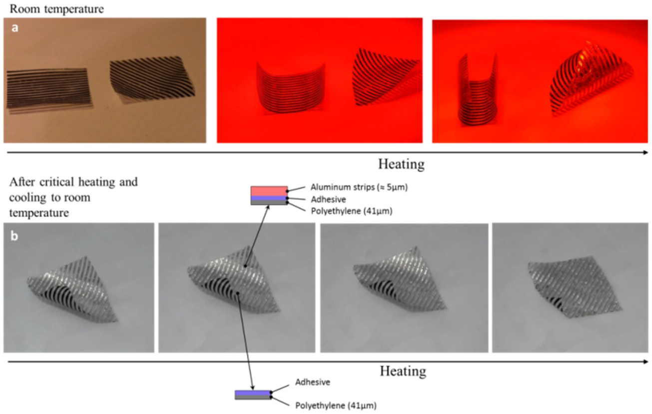

Figure 1. a) Transformation of two multilayer anisotropic materials (2D-to-3D shape) before the critical temperature level, b) Reverse transformation of the multilayer material (3D-to-2D shape) as the temperature increased, after the critical temperature level (the specimen in Fig. 1b is different from the specimen in Fig. 1a).

The first major step was the fabrication of the 2D sheet. A two-component adhesive for low-surface-energy plastics was used and applied to the oriented PE, Fig 2a (any adhesive for low-surface-energy plastics can be used). A mask was attached under pressure over an aluminium film using a hot rolling press at 180 ºC in order to form the strips at the desired directions, Fig 2b. The masked aluminium film was pressed together with the bilayer material using a cold rolling press, Fig. 2c. Then, the aluminium strips were formed using a chemical etching technique in which the masked aluminium strip was immersed in a ferric chloride solution at 40 ºC for < 20 min. The final multilayer material was cleaned and removed from the flat tool using acetone, Fig. 2d. The second major step was to form the 3D shape at room temperature. After a critical temperature level (T ≈ 95 ºC), the thermoplastic material come close to the melting temperature; then, the stiffness of the oriented layer was almost nullified (Fig. 2e) and the multilayer material transformed to a flat sheet. The external temperature may be applied either uniformly or locally. At this stage, the material could not react to the external temperature. During the cooling phase, the thermoplastic layer regained its stiffness;

during its contraction, deforms the multilayer material. During the cooling phase, the shape of the material transformed to a different 3D shape. Therefore, the initial shape of the material is a 3D geometry at room temperature, Fig 2f. The material remains functional and may be transformed from a 3D shape to a 2D shape or to a different 3D shape using the anisotropy of the layers and the mismatch in the coefficient of thermal expansions between the layers.

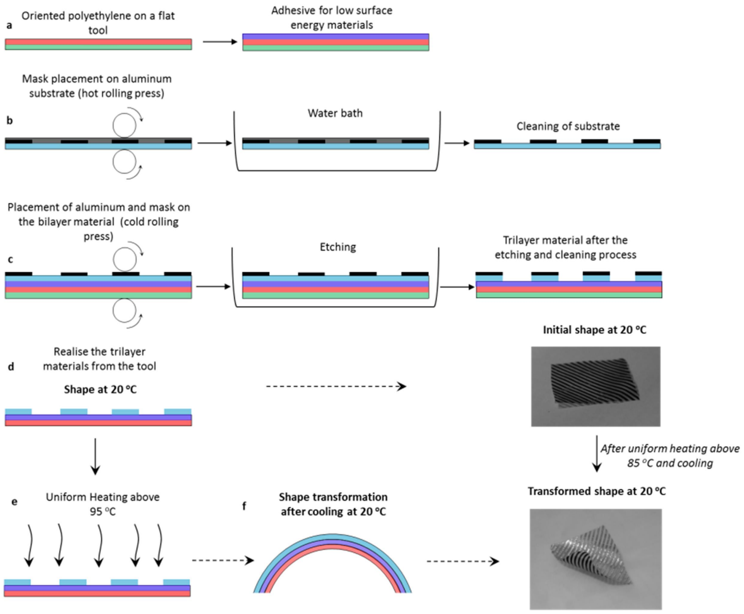

Figure 2. Fabrication process of the bioinspired materials. a) Fabrication of the bilayer material over a flat tool, (b) Mask placement over the aluminium film using a hot roll press and cleaning, c) Cold roll pressing of the different layers and formation of strips using etching techniques, d) Cleaning and demoulding. e, f) Transformation after the critical temperature level and 3D-shape formation after cooling at room temperature.

In addition, a second fabrication process will be presented. In this case, we followed exactly the same steps in order to fabricate the 2D sheets (Figs. 3a and b); however, the process of forming the 3D shape is differed. The multilayer material was formed via hot roll pressing, Fig. 3c. In this case, the layers were simultaneously heated and joined to one another. The internal stresses during the heating and cooling stage transformed the shape of the material and, as a result, a 3D shape was formed, Figs. 3c and d.

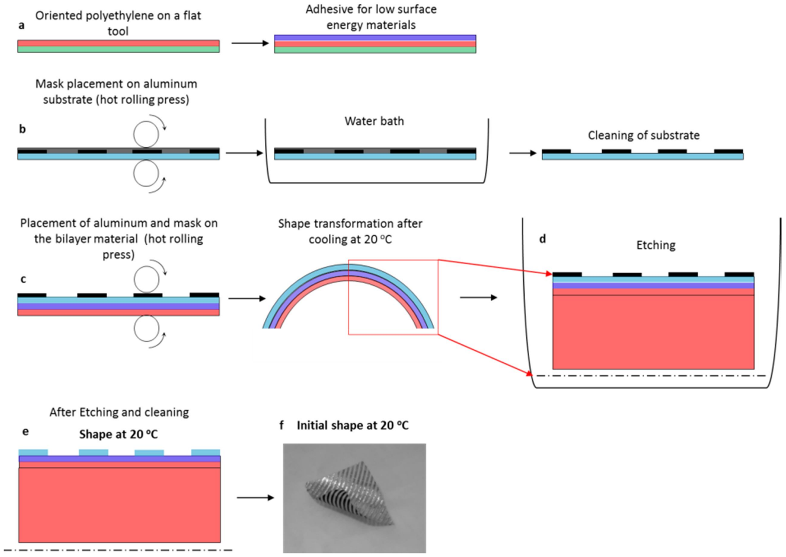

Figure 3. Fabrication process of the bioinspired materials. a) Fabrication of the bilayer material over a flat tool, b) Mask placement over the aluminium film using hot roll press and cleaning, c) Hot roll pressing of the different layers and transformation of the 3D-shape after cooling at room temperature, (d) Etching of the aluminium strips. e, f) Final 3D-shape at room temperature.

Then, the aluminium strips were formed using a chemical etching technique in which the masked aluminium strip was immersed in a ferric chloride solution at 40 ºC for < 20 min. During the cooling phase, the shape of the material transformed to a 3D shape. Therefore, the initial shape of the material is a 3D geometry at room temperature, Figs. 3e and f.

Both fabrication processes cases can be used for the continuous production of small or large surfaces of different thicknesses.

3. RESULTS AND DISCUSSION

3.1. Curvature of the multilayer film during the entire fabrication process

Figure 4 presents the curvature of the anisotropic material during the different stages. The black circles present the radius of the curvature of the multilayer material as the temperature increases. The flat shape of the multilayer material (Step 1, Fig. 4) transformed to a convex 3D shape owing to the CTE mismatch (Step 2, Fig. 4). At a steady-state temperature the multilayer material remains curved. After a critical temperature level has been exceeded (T ≈ 95 ºC), the modulus of elasticity of the polymeric substrate becomes nullified and the radius of the curvature of the multilayer material is maximised. The multilayer material folds and is transformed to a flat shape (Step 3, Fig. 4). During the cooling stage, the shape of the material transforms to a 3D shape with a negative curvature. Thus, the initial shape of the multilayer film is a 3D concave shape at room temperature (Step 4, Fig. 4). The radius of the curvature during the cooling stage differs from the radius of curvature during the first heating stage. This occurs because of the small temperature differentiations (non-uniform critical steady-state temperature) and the altered thermo-mechanical properties of the multilayer material after the critical heating. After the cooling phase, the initial shape of the material is a 3D geometry and the material is responsive to the temperature stimulus.

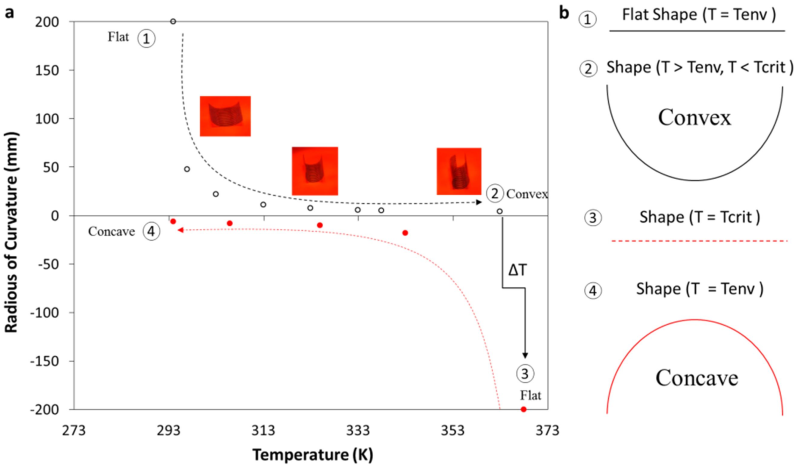

Figure 4. a) Measured radius of curvature of a multilayer material during the first heating stage and during the cooling stage, b) Schematic representation of the shape transformation for the entire fabrication process.

3.2. SEM characterisation after the fabrication process

A specimen was examined through SEM (scanning electron microscope, JEOL 6300) after the fabrication process. The scope of this characterisation was to evaluate the performance of the anisotropic multilayer material after the critical temperature of the fabrication process.

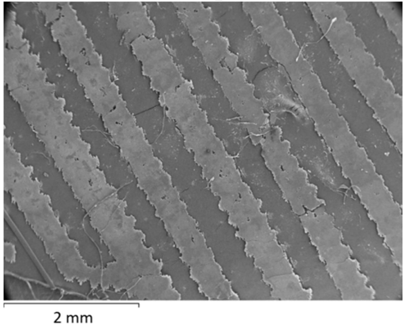

Figure 5. Crack formation of a multilayer film after the critical temperature level has been reached using SEM.

During the transformation of the material from 2D to a 3D shape and from a 3D convex shape to a 3D concave shape, failures can be formed due to the interlaminar failure between the strips and the substrate (PE), and buckling effects may occur, Fig. 5. These failures maybe eliminated using thicker aluminium strips and lower cooling rates [15]. Moreover, stronger adhesives would result in more durable materials.

3.3. Demonstration of a complex 3D-to-2D shape.

The scope of this section is to demonstrate a proof of concept and to present the capability of the multilayer materials to transform from a programmed 3D complex shape to a 2D- or to a different complex 3D shape. The multilayer material has been programmed to transform its initial geometry to a 3D geometry at room temperature. Figure 6a illustrates a "petal" that transforms from a 2D shape to a 3D shape when heated, whereas Fig. 6b presents a "petal" that transforms from a 3D shape to a 2D shape as the temperature increases.

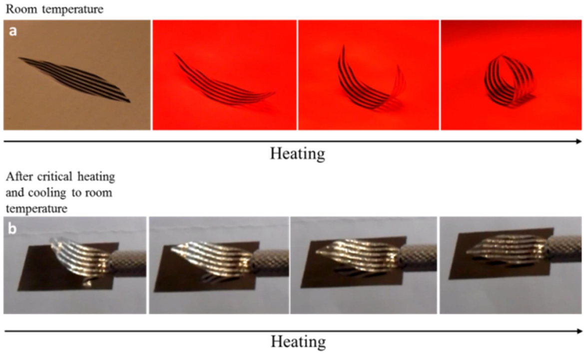

Figure 6. a) Petal transformation during the heating stage and b) petal transformation during the heating stage after the critical heating temperature has been reached.

Two different multilayer materials were assembled in order to mimic and fabricate the shape of a flower. Figure 7 depicts a thermographic image during the transformation of the formed "flower" as the temperature increases from room temperature (Fig. 7a) to a higher temperature (Fig. 7c). The shape of the flower returns to its initial 3D-shape at room temperature. In this particular case, one additional layer of silver has been applied to the external surface of the multilayer material (over the aluminium strips). Moreover, the thickness of the oriented PE and the metallic strips determine the rigidity and the sensitivity of the multilayer materials according to the application.

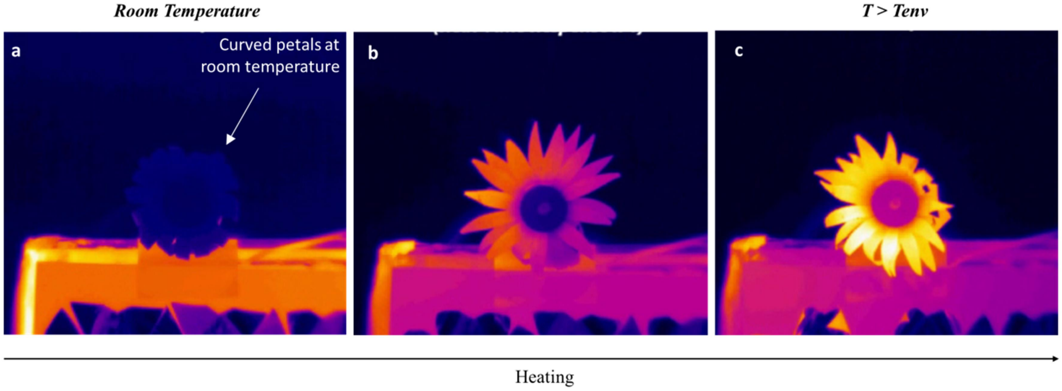

Figure 7. Transformation of a 3D complex shape as the temperature rises from room temperature to a higher temperature level.

4. CONCLUSIONS

The shape transformation of the multilayer anisotropic material can be triggered under a temperature stimulus. The initial programmed 3D shape of the multilayer material remained functional and could transform to another shape under temperature stimuli because of the CTE mismatch and the anisotropic properties of the multilayer film. By controlling the direction, and the distribution of the strips and the cooling rate, we designed a complex 3D shape that could be transformed reversibly to another shape as the temperature increased. Despite the fact that the multilayer material remained functional after the critical temperature, very thin aluminium strips could not withstand the excessive compressive stresses during the transformation. This problem could be resolved using thicker aluminium strips and stronger adhesives [15]. The proposed responsive multilayer anisotropic materials can be manufactured by employing etching and conventional techniques as per the suggested fabrication steps and by considering the material properties, the degree of anisotropy of the materials, and the direction of the layers. This approach bypasses the use of complex tools to form the geometry of the material in 3D space. A plethora of applications can incorporate the proposed materials and fabrication processes in order to programme and create a responsive 3D-shape material at room temperature that alter its geometry reversibly without the use of complex tools.

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