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Auxetic composite made with multilayer orthogonal structural reinforcement

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Abstract

Auxetic composites are non-conventional materials with negative Poisson's ratio (NPR). They have received great attention in recent years due to their particular properties and high application potential in different areas. In this study, a novel kind of auxetic composite was proposed and fabricated via an injecting and foaming process by using multilayer orthogonal auxetic structure as reinforcement and polyurethane foam as matrix. The NPR effect and mechanical behavior of the composite under quasi-static compression were investigated and compared with those of the pure polyurethane foam and non-auxetic composite. The results obtained show that the auxetic composite fabricated has an obvious NPR effect and behaves more like a damping material with a large range of deformation strain. The study has provided a simple way to design and fabricate auxetic composite materials by using suitable arrangement of reinforcement materials in a multilayer orthogonal structure.

Keywords: auxetic composite, multilayer orthogonal structure, negative Poisson's ratio, compression

1. Introduction

Auxetic materials are those having negative Poisson's ratios (NPR). They expand in the transverse and/or thickness direction when subjected to tensile stretch in the longitudinal direction, and shrink when compressed in a perpendicular direction, which results in a unique feature, that is, the materials can concentrate themselves under the compression to better resist the loads [1]. This particular feature in combination with other improved properties such as shear resistance [2-3], indentation resistance [4-5] and fracture toughness [2,6], has made auxetic materials very attractive for many potential applications, such as automobile [7], aerospace and defense [8], sensors [9] and protection, etc.

Auxetic materials have been studied for more than two decades. Since the first auxetic polyurethane foam was reported in 1987 [10], a number of auxetic materials have been proposed and fabricated based on different auxetic structures. The examples include auxetic polymeric foams and micro-porous polymers [6, 11], auxetic fibers [12-14] and fabrics [15-18], auxetic honeycombs [19] and composites [20-23], etc.

As a particular kind of auxetic materials, auxetic composites have received great attention in recent years. Alderson et al. [20] had reported that auxetic composite materials could be fabricated through two approaches. The first approach was to produce the fiber-reinforced composite laminates by using non-auxetic materials [20, 24-26]. The route was to use off-the-shelf pre-preg structure (angle ply) which gives specific stacking sequences to produce auxetic effects. Using this method, the composite laminates could be designed to have in-plane or out-plane negative NPRs.

However, the requirement for the auxetic composite laminate was that the individual ply materials should be highly anisotropic [20], which indicated that carbon fiber/epoxy resin [26-27] was more suitable choice than others [28]. The second approach was to use auxetic reinforcements. The auxetic fibers [14] embedded in the composite as the reinforcement would prevent the occurring of fiber pullout because the fiber would get fatter when stretched, leading to self-locking of auxetic fibers into the matrix. Composite using two-layer woven fabric made of double-helix yarns as inherently auxetic phase was manufactured to be auxetic with an approximate Poisson's ratio of -0.1 [29]. The shortcoming of this kind of composite was that the matrix silicone rubber gel seriously constrained the strong auxetic property of double-helix yarn with a Poisson's ratio of -2.1. Hou et al. [21] suggested another kind of composite structure with isotropic NPR by randomly including re-entrant triangle elements into a matrix. The Poisson's ratio of the composite structure could be tailored by changing the geometrical features of inclusions and properties of components [22] and high difference in stiffness of the inclusions and the matrix material was a condition to obtain NPR of the composite [23].

In this study, multilayer orthogonal structural reinforcement derived from a 3D auxetic textile structure developed in our previous studies [30, 31, 32] was employed to fabricate auxetic composite with compressible matrix. Auxetic effect and mechanical behaviors of the composite under quasi-static compression tests were analyzed and compared with non-auxetic composite made of the same materials, but with different arrangement of reinforcement in multilayer orthogonal structure. It is expected that such a study would provide direct insights into the design and fabrication of auxetic composite materials by using multilayer auxetic orthogonal structure as reinforcement.

2. Experimental

2.1 Multilayer orthogonal auxetic structure

Multilayer orthogonal auxetic structure used in this study was derived from a 3D textile structure developed in our previous studies [30, 31] by eliminating stitch yarns in the thickness direction of the structure because the stitch yarns that are used to bind all the layers of weft and warp yarns together when fabricating reinforcement part contribute a little to the compression properties of composite [32]. As shown in Fig. 1 (a), the structure consists of two parts of reinforcements in the x direction and y direction, respectively. Suppose that the reinforcement in the x direction is material A and the reinforcement in the y direction is material B, A and B are laid up alternately like wood pile. The cross-sections of the structure in y-z plane and x-z plane are shown in Fig. 1 (b) and (c), respectively. From the cross-section in y-z plane, it can be seen that material A is arranged in the same way in all the layers. While, from the cross-section in x-z plane, it can be seen that material B is arranged differently and a half-spacing shift exists between even layers and odd layers. Due to this particular arrangement, material A will be bent under action of material B when the structure is subjected to compression in the z direction, which leads to the contraction of the structure in the x direction (Fig. 1 (d)). This deformation behavior is very similar to that of the re-entrant hexagonal structure which is the most commonly used structure for auxetic materials. However, in the y direction, as material A is arranged in the same way in all the layers, material B will keep straight under compression and the length of the structure will keep unchanged. As a result, the multilayer orthogonal auxetic structure will have NPR in the x direction and zero Poisson's ratio (ZPR) in the y direction under compression.

To arrange the material A and material B to form a multilayer orthogonal auxetic structure and to facilitate the subsequent composite manufacturing process, a mold as shown in Fig. 2 (a) was first fabricated with stainless steel according to the geometrical parameters of the structure and the cross-sectional size of reinforcement materials selected. To let the structure easily shrink in the x direction and keep unchanged in the y direction under compression, the flexible polyester filaments were selected as material A and the rigid hollow ABS plastic tubes were selected as material B. The type of polyester filaments selected was of high-strength with low-shrinkage, which were provided by Shandong Helon Polytex in China. Their properties are as follows: material density=1.38g/cm^3; yarn linear density=1670dtex (456f); fracture stress=345.29MPa; fracture strain=4.63%; elastic modulus=12.77GPa. The ABS tubes were provided by Formosa Chemicals and Fibers Corporation and their properties are as follows: material density=1.05g/cm^3, diameter=3mm, elastic modulus=2.2GPa, bending modulus=28GPa, Poisson's ratio=0.394, Vicat softening point=103°C, heat deflection temperature=94°C.

According to the previous studies [30, 31], in addition to the cross-sectional size of reinforcement materials, the shifting distance L_0 of ABS tubes in the x direction as shown in Fig. 1 (c) also influences the NPR effect of the structure, and the decrease of L_0 can lead to an increase in the NPR effect. To get high NPR effect, L_0 should be set as low as possible. In this study, L_0 was selected as 4.5 mm. It was obtained by summing up two radiuses of ABS tubes (2*1.5mm) and walls thickness between the two neighbor slots of the mold (1.5mm). To decrease L_0, the wall thickness was selected according to the mechanical limitation of fabrication of the mold. Fig. 2 (b) shows a multilayer orthogonal auxetic structure fabricated by binding the polyester filaments (material A) around the steel fences in the x direction and inserting the ABS tubes (material B) between the slots in the y direction in the mold. All the polyester filaments and ABS tubes were kept in a straight form. To reduce the boundary effect under compression test in the z direction, a high number of layers of the structure should be adopted. Therefore, the multilayer orthogonal auxetic structure was fabricated with total 16 layers of reinforcements (8 layers for material A in x direction and 8 layers for material B in y direction). It was kept in the mold for the subsequent fabrication of composite.

2.2 Fabrication of composite

The composite was fabricated via an injection and foaming process. To do that, the multilayer orthogonal auxetic structural reinforcement together with the above stainless steel mold was first put into a metal mold, and then the uniformly-mixed polyurethane formulation was injected into the reinforcement by hand injection molding. The polyurethane formulation was composed of components M and N, where M was industrialized MDI Wannate PM-400 provided by Wanhua Chemical

Group Co., Ltd. in China, and N was a mixture of polyether polyols 330 provided by Jiangsu Haian Petrochemical Plant in China, dibutyltin dilaurate provided by J&k Scientific in China, α-Propylene glycol provided by Sinopharm Chemical Reagent Co., Ltd. in China, dimethyl silicone provided by Sinopharm Chemical Reagent Co., Ltd.in China, 1,4-Diazabicyclo[2.2.2]octane provided by J&k Scientific in China, deionized water provided by Sinopharm Chemical Reagent Co., Ltd. in China, dichloromethane provided by Sinopharm Chemical Reagent Co., Ltd. in China, and flame retardant XF-680 provided by Shanghai Xufan Chemical in China. All chemical materials were of analytical grade and used as received without further purification. After the foaming process had finished, the composite was cured in an oven at 90°C for 2 h. After de-molding, the obtained composite was cut to a specific size for the subsequent quasi-static compression tests. The picture of an auxetic composite sample is shown in Fig. 3 (a). For the comparison, the pure polyurethane foam and non-auxetic composite were also fabricated, as shown in Fig. 3 (b) and (c). As shown in Fig. 3 (c), the non-auxetic composite was fabricated with the same materials and same volume fraction of reinforcements, but with different arrangement of ABS tubes in the multilayer orthogonal structure, in which no shifting existed among the ABS tubes layers. The density of the pure polyurethane foam measured was to be 0.10g/cm^3, while the auxetic composite and non-auxetic composite were measured to have the same density of 0.27g/cm^3.

In the preliminary study, the composite was made with all hollow ABS tubes. However, a problem was found during the compression test. As shown in Fig. 4, the hollow tubes located in the top layer of the composite collapsed under high compression loads. In Fig. 4, the tubes collapsed are marked by red circles. There is no doubt that the collapse of tubes will directly weaken the mechanical performance of auxetic reinforcement thus lead to the failure of the whole composite. To avoid the easy collapse of ABS hollow tubes under high compression and to facilitate the measurement of the transverse strain, the solid ABS tubes with better compression resistance (Rockwell hardness R-110, ASTM D785) were used to replace the hollow tubes in the top layer, bottom layer, two sides of the auxetic composite and the top layer, bottom layer of the non-auxetic composite in this study.

2.3 Quasi-static compression tests

In order to assess the NPR effect and mechanical behavior of the auxetic composite produced, the quasi-static compressive tests were performed according to the ASTM D 1621 standard using an Instron 5900 testing machine installed with two 150mm compression circular plates, as shown in Fig. 5. The tests were carried out at laboratory temperature (25±3°C) and relative humidity (60±5%). The dimensions of specimens were cut as 100mm×98mm×28mm, and the compression rate was set at 2 mm/min. During the compression test, the vertical compression displacement of specimen was recorded directly by the machine. To measure the lateral displacement of specimen, a camera with a timer shot function placed at a distance of 50cm from the front face of specimen was used to synchronously record the lateral size change of the specimen. Eight points were marked with black color in two sides of each sample to facilitate recording deformation information during compression tests. To minimize the boundary effect of the top and bottom compression plates, only the displacement in the central two layers of the composite was used and the mean value of these two distances was adopted to calculate the lateral strain of the composite. The initial distance between the two sides' marked points was first measured before compression. During each compression test, thirty photos were taken at a time interval of 10 seconds. The distances of the marks in the photos were measured by a screen ruler to calculate the strain of composite in the lateral direction. Three samples were tested for each composite and the mean value was used to calculate the compressive strain and lateral strain. After the compressive strain ε_z and the lateral strain in the x direction ε_x were obtained, the Poisson's ratio of the tested sample ν_{exp} was calculated from Equation (1).

$$\nu_{exp} = - \varepsilon_x / \varepsilon_z \quad (1)$$

3. Results and discussion

3.1 NPR effect

The lateral deformation of the auxetic composite and non-auxetic composite during the quasi-static compression tests are shown Fig. 6 (a) and (b), respectively. From Fig. 6 (a), it can be seen that the auxetic composite laterally contracts under compression. This means that the NPR effect is achieved with auxetic composite. Due to the constraint of the compression plates, the top side and bottom side of the composite cannot contract under compression. As a result, the two lateral sides of the composite are not kept straight and indents with the shape of a crescent are formed. It is evident that the increase of reinforcement layers in the composite structure could reduce this boundary effect. Different from the auxetic composite, the non-auxetic composite does not laterally contract under compression. As shown in Fig. 6 (b), as the ABS tubes are arranged in the form of the vertical lines, a lateral deformation towards the right side takes place under compression, resulting in a collapse of the composite structure. The similar phenomenon was also reported in [33]. The above analyses indicate that although auxetic and non-auxetic composites were made with the same materials and the same volume fraction of reinforcement, their deformation behaviors are completely different because of different arrangements of reinforcements in the orthogonal structure.

The values of Poisson's ratio for the auxetic composite as a function of the compression strain calculated from the experimental results according to Equation (1) are shown in Fig. 7. It can be seen that the NPR value varies with the compression strain. From compression strain 0 to 15%, the Poisson's ratio of the composite almost approaches zero. To verify whether the quasi-zero Poisson's ratio (ZPR) phenomenon is caused by the unique configuration of the auxetic composite with solid tubes on the edges, the auxetic composite with all hollow tubes was compressed and its Poisson's ratio versus compressive strain curve was also shown in in Fig.7 for comparison. From Fig. 7, it can be seen that the curves of two types of composites have similar rising tendency and no difference exists from 0 to 15% of compression strain between them. This means that the ZPR effect of auxetic composite in the initial stage is not generated by the use of the rigid tubes on the sides. In fact, this phenomenon mainly originates from the difference between the thicknesses of the composite and its reinforced multilayer orthogonal auxetic structure as well as quasi-zero Poisson's ratio behavior of the foam self-fabricated in this study. According to the measurement result, the thickness of the auxetic composite fabricated was 28 mm, while the thickness of the reinforcement structure constructed by 8 layers ABS tubes and 8 layers polyester yarns is almost 24 mm. Hence, the difference of 4 mm just corresponds to about 15% of compression strain of the composite. It should be noted that this difference in thickness is produced by the expansion of polyurethane foam during the foaming process. As shown in Fig. 1, in the multilayer orthogonal auxetic structure, the polyester filaments located in the x direction closely contact with ABS tubes located in the y direction and no gaps exist at the contact points. However, due to the expansion of polyurethane foam during the foaming process, the polyester filaments in the composite structure cannot be kept in contact with the ABS tubes. Since the voids exist in the polyurethane foam and the small gaps exist between the polyester filaments and ABS tubes, the deformation of the composite in the initial stage of compression mainly comes from the compression of the polyurethane form. As the measured Poisson's ratio of self-made pure polyurethane foam is zero from 0 to 7.4% of compression strain and only has a maximum value of 0.06 at the strain of 15%, as shown in Fig. 8, meanwhile the polyester filaments are still kept straight within 15% of compression strain and its high tensile modulus property would help prevent the bulge of foam matrix, the lateral size of the composites is almost kept unchanged, producing a quasi-zero Poisson's ratio of the composites from 0 to 15% of compression strain. However, when the compression strain exceeds 15%, the

Poisson's ratio of the composite is getting negative. It can be seen from Fig. 7 that the NPR value nearly linearly increases from 15% to 50% of compression strain and reaches a maximum value of -0.105 at around 50% of compression strain. After 15% of compression strain, the foam is gradually compressed to a near-densified state and the polyester filaments get in contact with the ABS tubes. With progress of compression, the polyester filaments will be bent under compression loads transmitted from the ABS tubes, causing the lateral contraction of the composite. As the bending effect of the polyester filaments increases with the increase of the compression strain, the NPR value increases with the increase of the compression strain. However, after 50% of compression strain, the NPR value starts to reduce. In this compression stage, the foam has completely been compressed to a densified state. Meanwhile, the cross-sections of hollow ABS tubes in circular form also start to be deformed into flattened form. Therefore, further compression will cause the increase of the lateral size, resulting in a reduction of the NPR effect.

The NPR effect of the auxetic composite is mainly caused by the unique structure of its skeleton. Thus, the Poisson's ratio of the auxetic composite could be theoretically calculated through a geometrical analysis of the multilayer orthogonal structure. Based on the geometrical model proposed in our previous studies [30], as shown in Fig. 8, the Poisson's ratio of the auxetic composite and compression strain could be theoretically calculated using Equations (2) and (3), respectively.

$$\nu = -\frac{\left(L-L_0\right)/L_0}{\left(H-H_0\right)/H_0} = -\frac{2R*\left(\sin\theta-\theta\cos\theta\right)+L_0*\cos\theta-L_0}{2R*\left(\cos\theta+\theta\sin\theta\right)-L_0*\sin\theta-H_0}*\frac{H_0}{L_0} \qquad (2)$$

$$\varepsilon = \frac{H_0\text{-}H}{H_0} = \frac{H_0\text{-}2R\left(\cos\theta + \theta\sin\theta\right) + L_0 * \sin\theta}{H_0} \tag{3}$$

where v is the Poisson's ratio of the composite; ε is the compression strain; R is the radius of the ABS tube; θ is the inclination angle of the straight segment of the polyester filament; L and H are the horizontal and vertical distances of the central points between the two adjacent ABS tubes, respectively; L_0 and H_0 are the initial values of L and H.

The solid curve in Fig. 7 was drawn from Equations (2)-(3) using the following given geometrical parameters of the multilayer orthogonal structure: R = 1.5 mm, L_0 = 4.5 mm and H_0 = 2R = 3 mm. From the compression strain 15% to 50%, it can be observed that the theoretical calculation results are very close to but a little bit higher than the experimental ones. The reason is that the NPR effect of the skeleton decreases when confined by the matrix foam in the composite.

3.2 Compression behavior

The compression curves including the stress versus strain curves and specific stress (stress divided by density) versus strain curves of the pure foam, auxetic composite and non-auxetic composite are shown in Fig. 9 (a) and (b), respectively. It can be seen that the compression curves of these three materials are very different. As shown in Fig.9 (a), the pure polyurethane foam, which is used as the matrix, has the lowest compression performance among the three materials when the testing conditions including the dimensions of specimens are kept the same. But if comparing the specific stress as shown in Fig.9 (b), it could be found that owing to its very low density, the pure foam has similar or even a little higher specific stress than the auxetic composite before the compression strain reaches 33.48%. However, after the compression strain exceeds 33.48%, the increase of the specific stress of the auxetic composite gets much faster than that of the pure foam. When the strain reaches 41.46%, the specific energy of these two materials becomes the same. After the strain exceeds the strain exceeds 41.46%, the same mass of the auxetic composite will absorb more the energy than that of the pure foam, indicating that the auxetic composite has better compression performance than the pure foam. The advantages of auxetic composite consists in its enhanced mechanical properties within an acceptable scope of the density and better impact resistance under multiple low-velocity impact, which will be studied in the future work.

Compared with composite materials, the pure foam behaves more flexible and has a larger deformation. The easy deformation of the foam under compression allows the auxetic composite to have high lateral shrinking and large compression strain if the reinforcements in the orthogonal structure can be easily deformed. As the arrangement of the ABS tubes in the multilayer auxetic structure allows polyester filaments to be easily bent, the auxetic composite has higher compression strain than the non-auxetic orthogonal structures.

As shown in Fig. 9 (a), the compression curve of the auxetic composite can be roughly divided into three stages. The first stage is from 0 to 15% of compression strain. In this stage, the compression stress linearly increases with compression strain. The second stage is from 15% to 35%. In this stage, as the increase of the compression stress is very slow, a plateau region is formed. The compression strain after 35% can be considered as the third stage. In this stage, the compression stress rapidly increases due to the densification of the foam and composite structure. This three-stage deformation mode shows that the auxetic composite behaves more like a damping material. While for the non-auxetic composite, it can be found that the plateau region does not exist in its compression curve. As shown in Fig. 9 (a), after a slow increase of the compression stress from 0 to 15% of compression strain, the compression stress rapidly rises until 18% of compression strain where the collapse of the whole material occurs. After this strain, the compression stress promptly declines. As shown in Fig. 3 (c), the ABS tubes are arranged in a form of the vertical lines in the non-auxetic composite structure. This arrangement makes the ABS tubes play a major role in bearing and transmitting load during the compression process, which results in a rapid increase of the compression stress and a lower deformation strain of the non-auxetic composite. This arrangement also makes non-auxetic composite stiffer than the auxetic composite. From above analyses, it can be concluded that the auxetic composite behaves like a damping material, while the non-auxetic composite behaves more like a stiffer material. The difference in their compression behaviors mainly comes from different arrangements of the ABS tubes in the multilayer orthogonal structure.

The formation of the plateau region in the compression curve and high deformation strain range implicates that the auxetic composite has better energy absorption capacity than the non-auxetic composite. The non-auxetic composite fails at 18 % of compression strain due to the collapse of the structure. At this strain, the energy absorbed for non-auxetic composite is 31.97kJ/m^3. The same energy can be absorbed by the same volume of auxetic composite at a compression strain of 38.36%. However, the auxetic composite does not fail at strain 38.36% and will continue absorbing energy till the ultimate collapse of the structure. This behavior makes auxetic composite more suitable than non-auxetic composite for being used as energy absorbing material.

4. Conclusions

A novel kind of auxetic composite was fabricated using multilayer orthogonal auxetic structure as reinforcement and polyurethane foam as matrix via an injecting and foaming process. Its NPR effect and compression behavior under quasi-static compression were analyzed and compared with the pure polyurethane foam and non-auxetic composite made with the same materials but with different multilayer orthogonal structure. According to the results obtained, the following conclusions could be drawn.

1. The NPR effect of a composite can be achieved by using suitable arrangement of reinforcements in a multilayer orthogonal structure. This provides a simple way to produce auxetic composite using conventional materials.
2. The auxetic composite fabricated with multilayer orthogonal auxetic structure demonstrates an obvious NPR effect under compression and this effect varies with the compression strain. A highest NPR -0.105 is obtained when the compression strain reaches around 50%.
3. The auxetic composite and non-auxetic composite have different mechanical

behaviors due to different arrangements of the ABS tubes in their reinforcement structures. The auxetic composite behaves more like a damping material with a larger range of deformation strain, while the non-auxetic composite behaves more like a stiffer material with a lower range of deformation strain. The non-auxetic composite fails at 18% of compression strain due to collapse of the reinforcement structure.

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