Designing composites with negative linear compressibility

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HIGHLIGHTS
• Two approaches have been developed to design new composite structures with negative linear compressibility.
• The effectiveness of the design approaches has been proved by hydrostatic compression experiments.
• The designed composites process negative linear compressibility at large deformation.
• The negative linear compressibility is dependent on the foam property, the reinforcement pattern, and the stiffness ratio.

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ABSTRACT
The phenomenon of negative linear compressibility has attracted much interest because of its unusual deformation features with many potential applications. However, the design and fabrication of materials and structures with negative linear compressibility are limited. In this paper, we proposed two approaches to designing and fabricating new composite structures with negative linear compressibility. The effectiveness of the proposed design approaches was validated experimentally by applying uniformly distributed pressure to all surfaces of bulk specimens. The deformation features, strain history, and the effective area reduction of the specimens were analyzed from the experimental data. The results clearly demonstrated the feasibility of the proposed designing and manufacturing approaches for realizing composites with negative linear compressibility.

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1. Introduction
Materials and structures with uncommon behaviours such as negative Poisson’ ratio, negative stiffness, negative compressibility and negative thermal expansion have attracted intense research interest due to a variety of potential applications [1–5]. Negative compressibility is the least studied among them. Compressibility is a measure of the relative volume change of a fluid or solid as a response to a pressure change. Volumetric compressibility determines the material volume change under applied uniform pressure, whilst linear compressibility defines a change along a specific axis of the material. Conventionally, materials and structures contract three-dimensionally under a positive surrounded uniform pressure. This behaviour is defined as the positive linear compressibility along three main axes. However, some materials and
structures exhibit an unusual behaviour under the application of pressure, resulting in an increase in dimension along one or two directions. This deformation feature is referred to as the negative linear compressibility (NLC) or negative area compressibility (NAC). Such properties need not be associated to unstable systems especially because the overall volume compressibility is always positive [6]. Fig. 1 represents the schematic side view of the behaviour of NLC and conventional materials under the application of uniform pressure. For a material with entirely conventional volume compressibility, the uncommon NLC property may be accompanied by a large positive linear compressibility along the perpendicular directions to the NLC axis. The positive volume compressibility is one of the requirements for stability of an unconstrained block of material. According to elastic theory, the lack of constraint is equivalent to a surface traction boundary condition. This stability condition implies that the elastic modulus tensor is positive definite [7]. Therefore it is concluded that the materials with NLC property could be stable, so the elastic modulus tensor of NLC materials could be positive definite. The negative volume compressibility (NVC) behaviour is forbidden in classical thermodynamics [3]. The elastic modulus tensor of NVC materials is not positive definite, so they are not stable, but it is possible to have NVC performance in a pressure-induced phase transition [7]. Lakes and Drugan [8] presented a foam with strain-dependent NVC, and a negative bulk modulus in a pre-strained foam was observed and interpreted by Lakes and Wojciechowski [7]. Furthermore, metamaterials have the potential to be designed to achieve NVC [9].

NLC behaviour was firstly observed in tellurium along a particular direction, and it was reported by Bridgman in 1922 [10]. Baughman et al. [3] explained a variety of potential applications of the NLC phenomenon such as the development of optical telecommunication systems, artificial muscles [11], infiltrated porous structures and a new generation of pressure sensitive sensors and actuators [3,12,13]. The NLC behaviour was discovered at the molecular level such as LnNbO4 [14], Ag3(CoCN)6 [15], KMn3(AgCN)2 [16] and a large giant NLC in the molecular framework of Zn[Au(CN)2] [12]. At the micro and macro levels, Baughman et al. [3] proposed a wine-rack mechanism to explain the NLC effect. In such a mechanism, it is assumed that the rods were rigid in both the axial and transverse directions. When the system is subjected to stress, the hinged joints enable the wine-rack system to deform or restore the original shape when the stress is removed. In other words, the system deforms solely through the changes in the angles between the rigid rods. The rods can only rotate in the plane of a 2D system that is parallel to the direction of applying load. This assumption is valid when this hinging mechanism is 3D. Also, it is assumed that the stiffness of the structure resulted from resistance to relative rotation of the rods at hinges.

Weng et al. [17] presented a list of crystals that exhibited NLC. Barnes et al. [18] proposed a tetragonal network of nodes connected by a network of beams with NLC behaviour. Grima et al. [19] presented a 2D system with a honeycomb structure which exhibited NLC behaviour. In the following, a 3D system with NLC behaviour was generalized from the elongated hexagonal dodecahedron as the 3D equivalence of the honeycomb. The detailed analysis was presented by Grima et al. [6] to assess different uncommon behaviours of negative Poisson’s ratio, NLC, and NAC. A new hexagonal truss system with specific geometric features and NLC behaviour was presented by Grima et al. [20]. This system deformed through non-equal changes in the lengths of the cell walls when deforming through a constrained angle stretching rather than flexure or hinging of joints. Recently Zhou et al. [21] presented three 3D cellular models which exhibited negative compressibility. The new proposed systems have the tunable compressibility which can be adjusted for different applications with required properties of NLC, NAC, zero and negative Poisson’s ratios.

There has been extensive work on materials and structures design using the composite techniques, covering various uncommon properties such as auxetic [22–34], negative stiffness [35,36] and negative thermal expansion inclusions [37,38]. The basic idea of the composite technique is to improve the properties of materials and structures through adding a second phase to them in the form of fibre element or particulates [39]. In this study, we developed the simple design approaches to manufacturing a new series of structures with NLC property based on the application of composite techniques.

NLC composite materials and structures are rare and so far have been found in a very limited range of applications. Baughman et al. [3] presented a composite material with NLC property that consisted of a framework network with a helical chain configuration in low-density porous solid. Weng et al. [17] investigated the 2D and 3D composite structures with a rhombus-like hinged framework inside the filler component. All the above mentioned NLC composites have interior reinforcements with a wine-rack topology which all the members of the reinforcement were joined. Therefore the NLC property arises from the function of hinged joints. Thus, the corresponding mechanism for NLC can be presented by a wine-rack model which is frequently used to quantify NLC properties.

The NLC materials and structures with discrete and non-wine-rack elements are very rare. Recently Miller et al. [40] presented some common materials particularly carbon fibre laminate composites that exhibited NLC. The carbon fibre composites consisted of discrete carbon fibres which reinforced a stable matrix of epoxy. Also, Barnes [41] reported that the responsible mechanism for NLC could be different from a wine-rack model. A generic helical chain structure with discrete and non-wine-rack element was developed with NLC property [41]. In this study, we developed a new composite structure with a discrete reinforcement component which exhibited NLC behaviour. For some specific medical applications such as wound management, specific products with NLC behaviour are required as the wound filler. To transmit the liquids during curing, the void size of the wound filler should be in a specific range, i.e. 200–500 μm. Due to the manufacturing capacity, it is very difficult to produce a large quantity of NLC materials in meters with such a small dimension in its microstructures. A combination of the foam-type material as the filler component and the reinforcement component with NLC behaviour is a possible solution. In such case, the size of voids of new composite structures is determined by the cell size of the filler material. The reinforcement component plays a dominant role in the NLC behaviour of the composite structure.

To solve those challenges mentioned above, we proposed the new design approaches to design two types of NLC composite structures, respectively, composite structures with a reinforcement component-like a framework topology and composite structures with a discrete

![Fig. 1. Comparison of the behaviour of the conventional material and the NLC material.](image-url)
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