

## Mechanical behavior of composite gel periodic structures with the pattern transformation

Jianying Hu<sup>1</sup>, Yuhao He<sup>1</sup>, Jincheng Lei<sup>1</sup>, Zishun Liu<sup>\*1</sup>  
and Somsak Swaddiwudhipong<sup>2</sup>

<sup>1</sup>International Center for Applied Mechanics, State Key Laboratory for Strength and Vibration of Mechanical Structure, Xi'an Jiaotong University, Xi'an, 710049, China

<sup>2</sup>Department of Civil and Environmental Engineering, National University of Singapore, 117576, Singapore

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**Abstract.** When the periodic cellular structure is loaded or swelling beyond the critical value, the structure may undergo a pattern transformation owing to the local elastic instabilities, thus leading to structural collapse and the structure changing to a new configuration. Based on this deformation-triggered pattern, we have proposed the novel composite gel materials. This designed material is a type of architectural material possessing special mechanical properties. In this study, the mechanical behavior of the composite gel periodic structure with various gel inclusions is studied further through numerical simulations. When pattern transformation occurs, it results in a different elastic relationship compared with the material at untransformed state. Based on the obtained nominal stress versus nominal strain behavior, the Poisson's ratio and corresponding deformed structure patterns, we investigate the performance of designed composite materials and the effects of the uniformly distributed gel inclusions on composite materials. A better understanding of the characteristics of these composite gel materials is a key to develop its potential applications on new soft machines.

**Keywords:** periodic structure; gel materials; inhomogeneous field theory; numerical simulation; pattern transformation

### 1. Introduction

Periodic cellular structure has been widely investigated for generating novel pattern transformation and special mechanical properties. When it is loaded or swelling beyond the critical value, this type of periodic structure exhibits structural instabilities (Ding *et al.* 2013, Hong *et al.* 2009, Kang *et al.* 2013, Li *et al.* 2011, Mullin *et al.* 2007, Shim *et al.* 2012), thus leading to structural collapse and the transformation to a new configuration. The structure with the new configuration displays certain special mechanical properties, such as negative Poisson's ratio (Barnes *et al.* 2012, Bertoldi *et al.* 2010, Theocaris *et al.* 1997). So it is imperative to study the influence of instabilities on global material properties. Many researchers show their interests in designing the structures of materials to achieve or expand the special functionality of materials.

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\*Corresponding author, Professor, E-mail: [zishunliu@mail.xjtu.edu.cn](mailto:zishunliu@mail.xjtu.edu.cn)

Evans group (Alderson *et al.* 2010) had modeled chiral and anti-chiral honeycomb material structures, and considered the deformation mechanisms responsible for auxetic functionality of the two structures. Besides the honeycomb structures, the other shapes of re-entrant structures (Barnes *et al.* 2012, Gaspar *et al.* 2011, Miller *et al.* 2011) or buckliball structures (Shim *et al.* 2012) have also attracted large numbers of researchers to study the pattern transformation of those structural materials, especially for studying the critical value of applied load for different pattern. Meanwhile, several researchers have studied mechanical properties of two and three-dimensional soft cellular structures through theory, simulations and experiments (Bertoldi *et al.* 2008, Bertoldi *et al.* 2010, Geus 2011, Mullin *et al.* 2007, Willshaw and Mullin 2012). In the application of structural materials, Jang *et al.* (2009), Li *et al.* (2012) combined pattern instability and shape-memory hysteresis for photonic switching. Several studies on the collapse of a void or hole in an elastomer under swelling and de-swelling were also carried out (Cai *et al.* 2010, Ding *et al.* 2013, Hong *et al.* 2009). The similar novel composite material is experimentally studied by combining the silicone rubber samples with jelly filling each of the holes (Mullin *et al.* 2013). Based on the novel deformation-triggered pattern, we have developed the novel composite gel structure material. The arrays of shuriken (or four-pointed star) gel inclusions, quadrate gel inclusions, convex octagonal gel inclusions and circular gel inclusions are filled into the periodic cellular structures. This designed material is a type of architecture material which can achieve special mechanical properties and display a new mechanical behavior.

In our earlier study (Hu *et al.* 2013), we have investigated how the shapes of voids affect the mechanical properties of porous material through numerical simulations. While in this study, from the obtained nominal stress versus nominal strain curves and corresponding deformed structure patterns, we try to expound the effect of uniformly distributed gel inclusions with various shapes on the characteristics of the novel composite materials. Meanwhile, the influence of the gel behavior on the mechanical properties of composite gel periodic structure with pattern transformation is also studied.

## 2. Modeling of the composite material

The composite soft material composed of two components has already been manufactured by Mullin *et al.* (2013). In their experiment, the square arrays of circular holes in silicone rubber sheet are filled with jelly. At same time, we have independently carried out modeling and simulation study on mechanical behavior of hydrogel inclusion composite materials (Hu *et al.* 2013). According to the previous study, we understand that if the holes are filled with a more rigid material than that of the matrix, the cellular structure will not lead to any pattern switch (Michel *et al.* 2007). Therefore, a much softer material needs to be placed in the holes to achieve pattern switch for composite material. Mullin *et al.* (2013) has discovered that the inclusion which has a Young's modulus of  $>1\%$  the bulk would suppress the pattern switch. Based on the work on the incremental modulus of gel (Liu *et al.* 2011), we plug the gel materials into the porous elastomers and discuss how the gel material effects on the critical strain. In this study, the numerical simulations of composite material under compressed load are carried out to study the deformation behavior of the novel material. In the modeling, the specific combinations of two materials are modeled as illustrated in Fig. 1 (a), (b), (c), (d). The size of the model as shown in Fig. 1(a) is 40 mm×40 mm, comprising a microstructure of a 10×10 mm<sup>2</sup> square arrays of shuriken gel of 3.635 mm length with 10mm center-to-center spacing both vertically and horizontally; The size of model

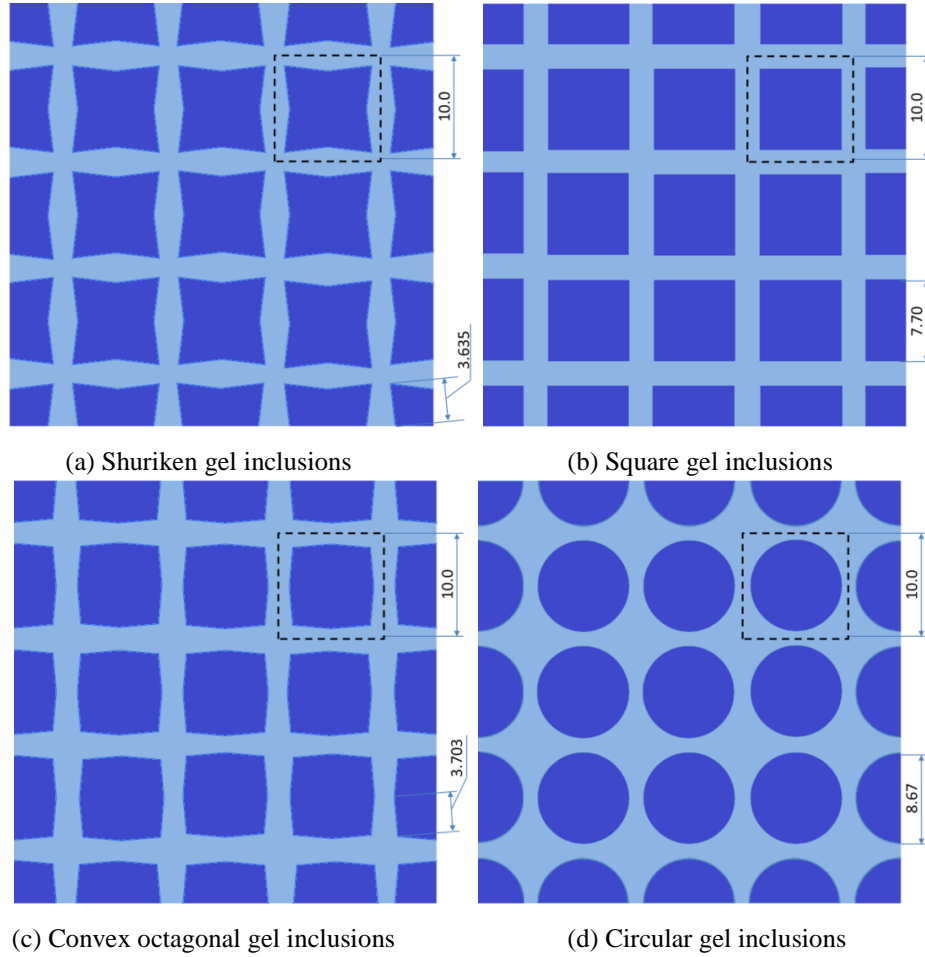


Fig. 1 Composite material structures with arrays of various gel inclusions. The gel fraction in four models shares the same value of 0.59

in Fig. 1(b) is 40 mm×40 mm with 10×10 mm<sup>2</sup> square arrays of square gel inclusions of 7.7 mm length with 10mm center-to-center spacing; The size of model in Fig. 1(c) is 40 mm×40 mm with 10×10 mm<sup>2</sup> square arrays of 3.703 mm length of convex octagonal gel with 10 mm center-to-center spacing; The size of Fig. 1(d) is 40 mm×40 mm, comprising a microstructure of a 10×10 mm<sup>2</sup> square arrays of circular gel inclusions of 8.67 mm diameter with 10mm center-to-center spacing. The volume fractions of gel in the four types of composite materials share the same value of 0.59.

## 2.1 Material properties

In the composite gel structural material, two different hyperelastic materials are used here, i.e., PSM-4 (Photoelastic Elastomer) for matrix and hydrogel for inclusions (Hu *et al.* 2013). Their energy densities are given in terms of the following two invariants  $I$  and  $J$  associated with deformation gradient  $\mathbf{F}$

$$I \equiv F_{ij}F_{ij}, J \equiv \det(F_{ij}) \quad (1)$$

For matrix material, a compressible neo-Hookean model (Lopez-Pamies and Castaneda 2004) is assumed. The strain energy  $W(\mathbf{F})$  form for neo-Hookean material in plane strain is

$$W(\mathbf{F}) = \frac{G}{2}[(I - 3) - 2\ln J] + \frac{\lambda}{2}(J - 1)^2 \quad (2)$$

where  $G$  and  $\lambda$  denote the standard Lamé moduli of the solid at zero strain. The material PSM-4 was modeled as nearly incompressible, characterized by  $k/G = 50$ , where  $k = \lambda + 2 \cdot G/3$ . From the early studies (Bertoldi *et al.* 2008, Mullin *et al.* 2007, Willshaw and Mullin 2012), the initial Young's modulus was given as  $E = 3.25\text{MPa}$ ,  $\nu = 0.49$ , so that  $G = \frac{E}{[2(1+\nu)]} = 1.1\text{MPa}$ .

The inclusion is assumed as gel material (Hong *et al.* 2009; Hong *et al.* 2008), with a strain energy  $W(\mathbf{F})$  given by

$$W = \frac{1}{2}NkT(I - 3 - 2\log J) - \frac{kT}{\nu} \left[ (J - 1) \log \frac{J}{J - 1} + \frac{\chi}{J} \right], \quad (3)$$

where  $N$  is the number of polymeric chains per reference volume, and  $\chi$  is a dimensionless measure of the enthalpy of mixing,  $kT$  is the temperature in the unit of energy and when at room temperature,  $kT = 4 \times 10^{-21}\text{J}$ . A representative value of the volume per molecule is  $\nu = 10^{-28}\text{m}^3$ . The two dimensionless material parameters  $N\nu$  and  $\chi$  are chosen appropriately, adopting the following 2 values,  $N\nu = 10^{-2}$  and  $\chi = 0.1$ .

## 2.2 Boundary condition

Just following our previous study, the composite material can be represented as a periodic array of representative volume elements (RVEs) with the aim of eliminating the boundary condition effects. Thus the numerical investigations are performed on periodic structures. Through extensive study for different models and compared with analysis of the full finite structure model, we also found that the RVEs results can exhibit an earlier switch (Mullin *et al.* 2007), i.e., boundary effects delay the structure transformation. The general form of periodic boundary condition can be expressed as (Berger *et al.* 2005, Xia *et al.* 2003)

$$u_i = \overline{S_{ij}}x_j + u_i^*, \quad (4)$$

where  $\overline{S_{ij}}$  are the average strain,  $u_i^*$  is the periodic part of the displacement components (local fluctuation) on the boundary surfaces, which is dependent on the applied global loads. The indices  $i$  and  $j$  denote the two-dimensional coordinate directions in the range of 1 to 2. A more explicit form of periodic boundary conditions, suitable for square RVE models can be derived from the above general expression. For a 2-D square RVE as shown in Fig. 2, the displacements on a pair of opposite boundary surfaces (with their normal along the  $x_j$  axis) are

$$u_i^{R+} = \overline{S_{ij}}x_j^{R+} + u_i^{*R+}, \quad (5)$$

$$u_i^{R-} = \overline{S_{ij}}x_j^{R-} + u_i^{*R-}, \quad (6)$$

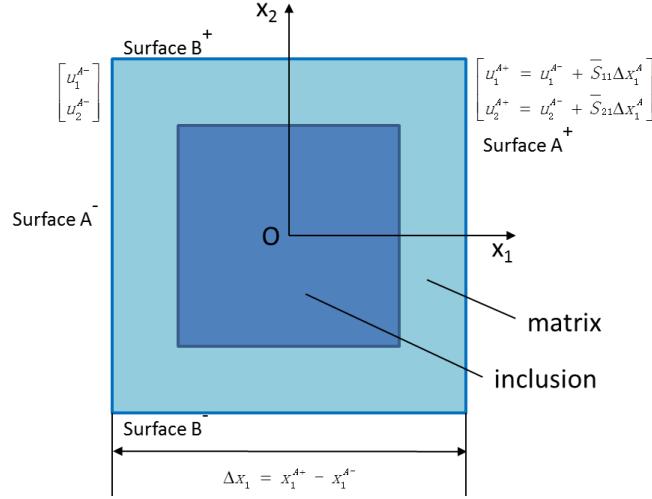


Fig. 2 Schematic diagram of a periodic composite RVE

where index “ $R+$ ” means along the  $x_j$  positive direction and “ $R-$ ” means along the direction on the corresponding surfaces  $A+/A-$ ,  $B+/B-$  (see Fig. 2 ). The local fluctuations  $u_i^{R+}$  and  $u_i^{R-}$  around the average macroscopic value are identical on two opposing faces due to periodic conditions of RVE. So, the difference between the above two equations is

$$u_i^{R+} - u_i^{R-} = \overline{S_{ij}} (x_j^{R+} - x_j^{R-}) = \overline{S_{ij}} \Delta x_j^R. \quad (7)$$

For any RVE model  $\Delta x_i^R$  is constant and this form of boundary conditions meets the requirement of displacement periodicity and continuity.

### 3. Pattern switching in composite material

When periodic elastomeric cellular solid is compressed, the array of pores undergoes an unstable transformation at a critical point (Bertoldi and Boyce 2008, Bertoldi and Boyce 2007, Bertoldi *et al.* 2008, Bertoldi and Gei 2011, Bertoldi *et al.* 2010, Jang *et al.* 2009, Kang *et al.* 2013, Mullin *et al.* 2007, Singamaneni *et al.* 2009a, Singamaneni *et al.* 2009b). Similar instabilities also trigger the transformation to the new configuration in the novel composite gel material (Hu *et al.* 2013). The numerical approach captures the mechanical behavior of composite material for exploring the effect of gel inclusions in various shapes. The property of gel material also greatly influences the pattern transformation of the composite materials.

#### 3.1 Stress versus strain behavior

The numerical results for the nominal stress versus nominal strain behavior of the four types of composite materials are displayed in Fig. 3. As we predicted, the behavior of the composite

material goes along the initial almost linear elastic behavior with a sudden change to a different elastic relationship. Fig. 3 also provides a direct comparison among the matrix materials with various gel inclusions. The transition point response to the pattern transformation in the composite materials and the patterns at the nominal strain of 0.2 are shown in Fig. 4. The gel inclusions of the novel composite material break their initial shapes and bifurcate into new shapes of vertical and horizontal directions alternatively.

Despite sharing the same value of gel fraction, the pattern transformation works differently in composite materials with gel inclusions of different geometric shapes. Fig. 5(a) depicts the  $20 \times 20 \text{ mm}^2$  microstructures for the four composite materials. The novel composite material is characterized by the length between center and re-entrant corner. The characteristic length gradually varies from 3.6 mm, 3.85 mm, 4.0 mm and 4.33 mm to describe the composite material with shuriken gel inclusions, square gel inclusions, convex octagonal gel inclusions and circular gel inclusions, respectively. Images of a square lattice of circular holes prior to loading and post-buckling are shown in Fig. 5(b) and (c). Emphasis is placed on the interstitial connectors which are considered as diamond-shaped units (light blue in Fig. 5(a); impassive yellow in Fig. 5(b) and (c)). At the turning point to the new configuration, much of the macroscopic deformation is observed to be accommodated by the rotation of the four matrix domains diagonally bridging neighboring inclusions (diamond-shaped units); these domains experience negligible strain but undergo large rotations. Neighboring diamond-shaped connectors rotate in opposite senses as a result of the buckling of the ligaments (non diamond-shaped domains in matrix material) and give rise to the pattern transformation for the composite material, the same as that of porous soft material in the experimental results of Willshaw and Mullin (2012). Then after transformation, due to that gel inclusion suppress the motivation of the matrix domains, the rotation leads to a different composite structure from the initial state. Obviously, the larger area of the diamond-shaped domains leads to the faster pattern switching. It can be inferred that the characteristic length has positive effect both on the modulus of elasticity before the buckling and the pattern transformation of composite material, thus reasonably explaining the curves in Fig. 3.

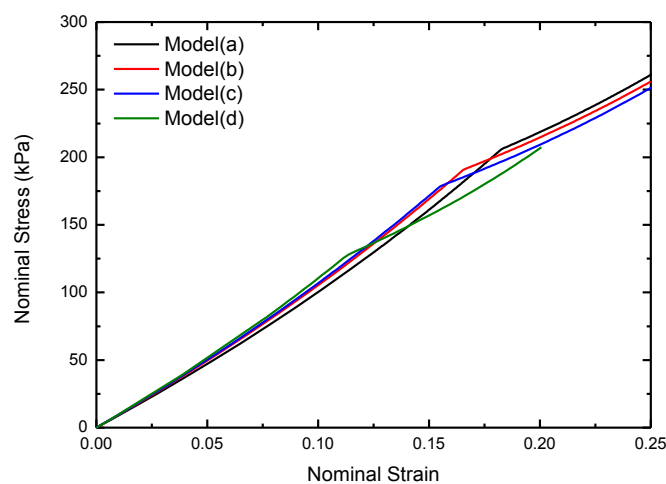


Fig. 3 The nominal stress versus nominal strain behavior of the four types of structures with various gel inclusions

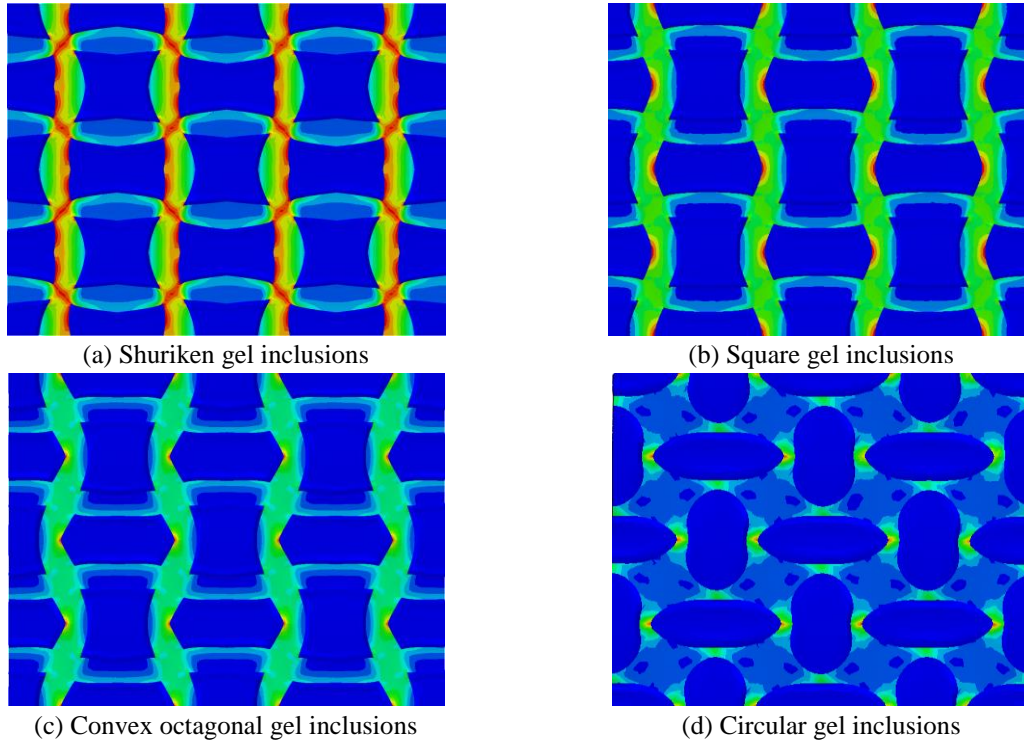


Fig. 4 The von-Mises stress distribution on transformed deformation pattern of the composite gel material structures at the nominal strain of 0.2

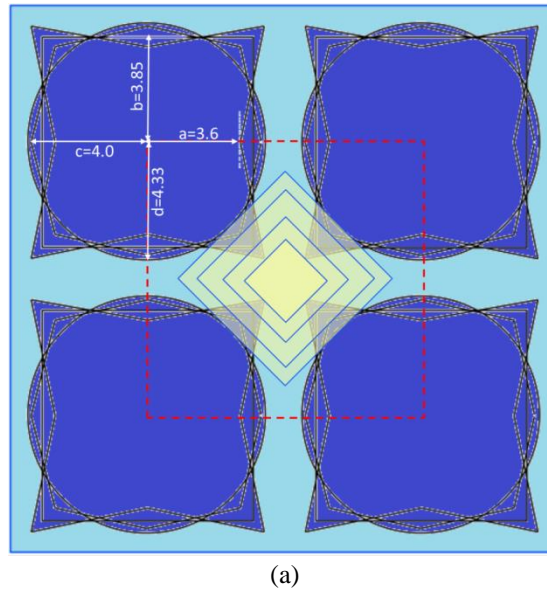


Fig. 5 A schematic diagram of microstructures for the composite materials with four geometrical gel shapes is given in (a). Figs. (b) and (c) show the Images of a square lattice of circular gel inclusions prior to loading and post-buckling case. Emphasis is placed on the interstitial connectors which are considered as diamond-shaped units (impassive yellow in (a) (b) and (c))

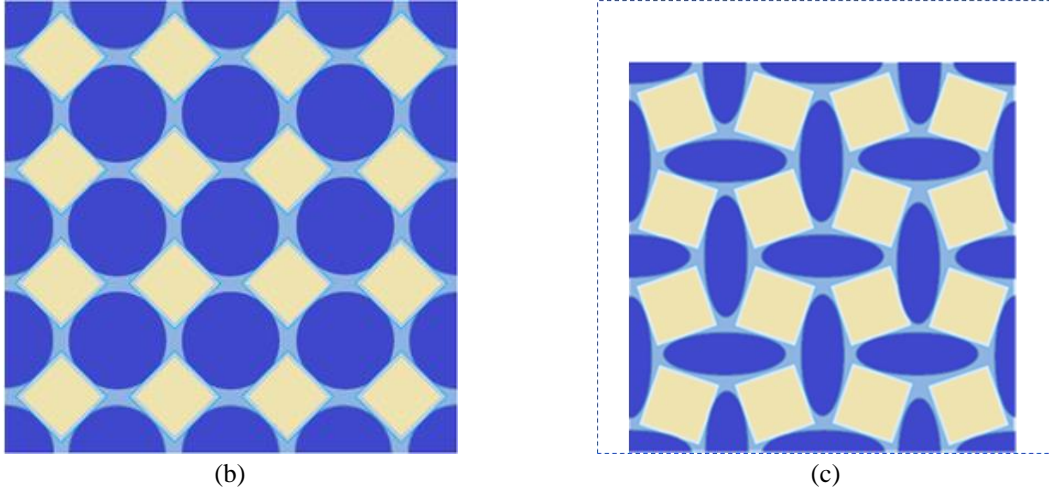


Fig. 5 Continued

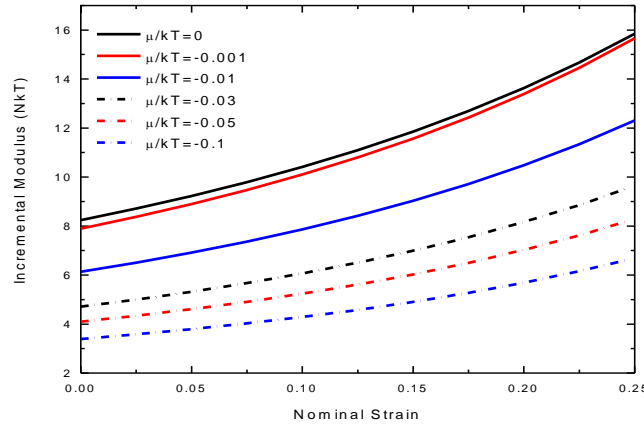


Fig. 6 Incremental modulus of gel varying with nominal strain for various initial chemical potentials

Besides the different shapes of gel inclusions, the property of gel inclusion also affects the properties of the composite material. As discussed in Section 2.2, we use gel mono-phase theory to simulate the gel inclusions. The initial state of gel inclusions in these novel composite materials is characterized by the free-swelling stretch  $\lambda_0$ , which is equilibrated in a solvent of chemical potential  $\mu$  as follows (Hong *et al.* 2009)

$$Nv \left( \frac{1}{\lambda_0} - \frac{1}{\lambda_0^3} \right) + \log \left( 1 - \frac{1}{\lambda_0^3} \right) + \frac{1}{\lambda_0^3} + \frac{\chi}{\lambda_0^6} = \frac{\mu_0}{kT} \quad (8)$$

When the composite material is compressed, gel inclusions are subjected to constraint deswelling, which is similar to the example case of a 1-D rod of a gel equilibrated in a solvent of chemical potential  $\mu$ , and subjected to a uniaxial stress  $S_1$  along the longitudinal direction. From the theoretical calculation of Liu *et al.* (2011), the reduced incremental modulus or tangent



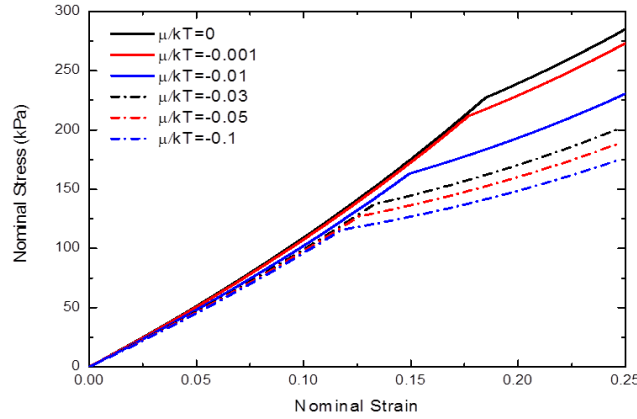


Fig. 7 Stress vs. strain behavior using the RVE model with array of square gel inclusions in an elastomeric matrix various for initial chemical potentials

stiffness of gel is a function of current deformation and stretch (current chemical potential), as illustrated in Fig. 6. According to the definition of incremental modulus of hydrogel, the value of the incremental modulus of gel increases as the stretch value decreases or the initial chemical potential increases.

We exploit gel subjected to different chemical potential, to fill the holes of porous elastic material to develop this novel composite material. Fig. 7 shows stress versus strain behavior using the RVE models with array of square gel inclusions in an elastomeric matrix various for initial chemical potentials.

We have observed several interesting properties of the new composite materials depicted in Fig. 7. Firstly, instead of perfectly linear elastic before the pattern switching, the material shows the increasing modulus because the gel inclusions becomes harder as the water in gel inclusions is squeezed out due to the applied load as shown in Fig. 6; so does the material after transformation. Secondly, the larger the initial chemical potential, the much more difficult the pattern transformation. It can be explained that the modulus of the gel increases as the initial chemical potential increases as illustrated in Fig. 6.

### 3.2 Poisson's ratio

Numerical and experimental studies show that the cellular structures with arrays of holes has negative Poisson's ratio when taking pattern switching (Bertoldi *et al.* 2010, Hu *et al.* 2013, Theocaris *et al.* 1997). Compared with the negative Poisson's ratio of structure with no gel inside (Hu *et al.* 2013), the pattern switching state exhibits a positive value of the Poisson's ratio in the composite material with square gel inclusions. Although the Poisson's ratio is positive for the gel composite materials, the tendency and shape of Poisson's ratio are the same as those of the porous materials studied by Bertoldi *et al.* (2010).

The difference may be attributed to the existence of gel inclusions. Without the gel inclusions, the volume of hole, or the area of the section of the hole, decreases under compression. While the gel inclusions is modeled as nearly incompressible material, the volume fraction of gel inclusions can not easily compressed just like the holes. That's the reason why the Poisson's ratio decreases

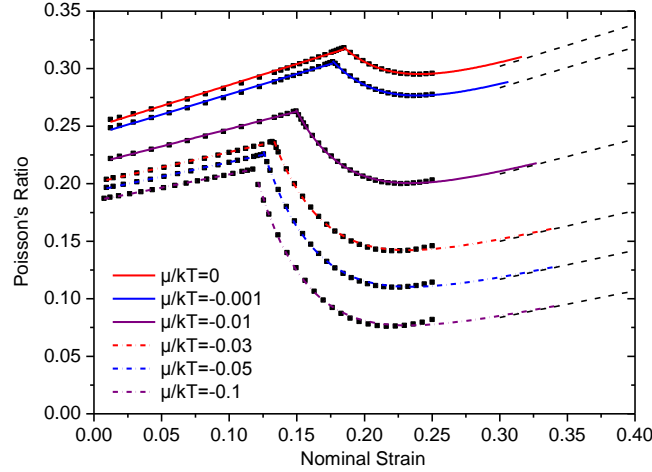


Fig. 8 Poisson's ratio as a function of nominal strain using the RVE model with array of square gel inclusions in an elastomeric matrix for various initial chemical potentials

but not reaches the negative value under compression.

The numerical results of Poisson's ratio are plotted as a function of nominal strain in Fig. 8 (data points) for slightly different values of chemical potential. A strong dependency on this parameter is evident. We find out that the Poisson's ratio of the composite material,  $\nu$ , decreases when the pattern transformation induced by the instability occurs. It is also striking that by simply decreasing the solvent chemical potential of the gel, composite material can be more easily reconstructed and the Poisson's ratio has a more sharp decline after switching point,  $\nu_c$ . Unlike the numerical results that lowest value of the asymptotic Poisson's ratio of the cellular porous structure was a constant (Bertoldi *et al.* 2010), the asymptote  $\nu_\infty$  that lowest value of the Poisson's ratio of composite material infinitely reaches numerically has the same slope as the fitted line of the Poisson's ratio before the pattern transformation. We observe that the results from the RVE simulations (data points) in Fig. 8 can be accurately regressed depicted as solid lines by exponentials of the form (Bertoldi *et al.* 2010)

$$\nu = \nu_\infty + (\nu_c - \nu_\infty) \exp \left[ -\frac{\varepsilon - \varepsilon_c}{\varepsilon_0} \right]$$

where,  $\nu_\infty$ , is the asymptote of Poisson's ratio values, the slope of which is the same as the fitted line prior to switching;  $\nu_c$  is the value of Poisson's ratio at the onset of the instability which occurs at a nominal strain;  $\varepsilon_c$ , and the characteristic strain of decay;  $\varepsilon_0$ , measures the speed of reaching the asymptotic value.

#### 4. Conclusions

We proposed the construction of novel composite materials by filling the gel inclusions into the periodic elastomeric cellular structures. This paper continues investigating the mechanical properties of the designed composite material. We hope that the mechanical properties of the novel

composite materials can meet its potential applications on future soft machines.

Numerical simulations of novel composite materials with various gel inclusions are carried out. The composite materials can be characterized by the length between center and re-entrant corner. The characteristic length has positive effect both on the modulus of elasticity before the buckling and the pattern transformation of composite materials, which may provide future perspectives for optimal design or serve as a fabrication guideline of the new gel composite materials.

It is observed that the internal microstructure of composite material greatly affects mechanical characteristics, so does the property of gel inclusions in composite material. The larger the initial chemical potential, the more difficult the pattern transformation. Meanwhile, the asymptote that lowest value of the Poisson's ratio infinitely reaches numerically has the same slope as the regressed line of the Poisson's ratio before the pattern transformation. The exponential forms are particularly well correlated.

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