Synergistic Energy Absorption Mechanisms of Architected Liquid Crystal Elastomers

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A unique rate-dependent energy absorption behavior of liquid crystal elastomer (LCE)-based architected materials is reported. The architected materials consist of repeating unit cells of bistable tilted LCE beams sandwiched between stiff supports. The viscoelastic behavior of the LCE causes the energy absorption to increase with strain rate according to a power-law relationship, which can be modulated by changing the degree of mesogen alignment and the loading direction relative to the director. For a strain rate of 600 s\(^{-1}\), the unit cell exhibits up to a 5 MJ m\(^{-3}\) energy absorption density, which is two orders of magnitude higher than the same structure fabricated from poly(dimethylsiloxane) elastomer and is comparable to the dissipation from irreversible plastic deformation exhibited by denser metals. For a multilayered structure of unit cells, nonuniform buckling of the different layers produces additional viscoelastic dissipation. This synergistic interaction between viscoelastic dissipation and snap-through buckling causes the energy absorption density to increase with the number of layers. The sequence of cell collapse can be controlled by grading the beam thickness to further promote viscous dissipation and enhance the energy absorption density. It is envisioned that the study can contribute to the development of lightweight extreme energy-absorbing metamaterials.

1. Introduction

Energy-absorbing materials are used in many daily and advanced applications for vibration isolation (e.g., in sports equipment and engine mounts), and impact protection (e.g., in helmets and armor). Many helmet and impact-absorbing materials dissipate energy through inelastic mechanisms, such as plastic deformation and fracture and fragmentation. However, these materials can become permanently damaged after one-time usage and are not suitable for repeated use. More recently, architected materials (or metamaterials) have been designed to trap energy through elastic buckling instabilities.\(^1\)\(^-\)\(^5\) This structural energy trapping mechanism is scalable and reversible, which makes the architected material reusable.\(^3\)

Nevertheless, the elastic energy trapping mechanism has a fixed energy absorption capacity regardless of strain rates.\(^3\)

It would be desirable to develop a reusable architected material that exhibits increased energy absorption over a wide range of strain rates for enhanced vibration and impact protection performance.

To achieve this objective, we hypothesized that the capacity for energy absorption of architected materials can be augmented by incorporating a rate-dependent material dissipation mechanism.\(^3,\)\(^6\)-\(^8\)

While the concept of architected materials is based on the interplay between material and geometry, most studies have focused on the nonlinear effects of mechanical instabilities rather than material inelasticity.\(^5\)

Recently, there were few studies that applied viscoelasticity to modulate the buckling pattern in multistable metamaterials. For example, Jang et al.\(^9\) demonstrated how strain rate-dependent mechanical metamaterials can be realized using bi-beams consisting of two laterally attached beams, where one is hyperelastic and the other is visco-hyperelastic.
The predictable switching of the buckling direction of the bi-beams is utilized to design lattice structures that can switch between auxetic and conventional behavior and that exhibit negative viscoelasticity. In addition, Dykstra et al. investigated the interaction between viscoelasticity and buckling instability by using a 3D-printed viscoelastic polymer to fabricate a metamaterial consisting of a 5 × 5 alternating square pattern of circular holes. They found that a geometric pattern change resulting from snap buckling could be delayed and even suppressed by increasing the loading rate. Finally, Che and coworkers developed viscoelastic multistable architected materials, where the sequence of snap buckling can be tuned by varying the temperature and beam thickness. However, little has been studied so far to investigate the synergistic interaction between viscoelasticity and architecture of metamaterials for enhancing energy absorption, while some investigators have leveraged rate-dependent frictional and surface interactions to increase energy absorption due to the synergistic interplay between the viscoelastic behavior of the LCE material and the snap buckling of the material architecture.

To fabricate the bistable architected LCE structures, we first synthesized main-chain LCEs using a two-stage thiol-acrylate reaction. A diacrylate mesogen, RM257 (1,4-bis(4-acyloxyloxy)-propoxybenzoyloxy)-2-methylbenzene), was used to form the liquid crystal phase. To connect the mesogens, a di-functional monomer (C3, 1,3-propanediolith) and a tetra-functional thiol monomer (PETMP, Pentaerythritol tetrais (3-mercaptopropionate)) were selected as a flexible spacer and a crosslinker, respectively (Figure 1a, Section S1, Supporting Information). During the first stage, a polydomain LCE was created by the thiol-acrylate Michael addition reaction. Next, the global arrangement of mesogens was further tailored by applying mechanical strain. After stretching a polydomain LCE, the photopolymerization reaction was used to establish crosslinks between the excess acrylate groups and fix the mesogen arrangement. Mechanical stretch causes the mesogens to rotate towards the loading direction and produce a soft stress response starting at 30% strain. At 90% strain, most of the mesogens have rotated towards the loading direction to complete the transformation of the polydomain to a monodomain. To investigate the effect of mesogen alignment on energy absorption, we prepared LCE sheets with various mesogen alignments by applying different mechanical strains: 0% (random alignment) and 90% (full alignment) before photo-crosslinking. We also investigated the effect of the alignment direction by cutting beams from the sheets parallel or perpendicular to the director. The beams were tilted and inserted into the stiff frame to create a bistable unit cell structure (Figure 1b, Section S2, Supporting Information).

2. Results and Discussion

The unit cells with different mesogen alignment with respect to the beam direction were subjected to uniaxial compression at different effective strain rates, defined as displacement rate divided by the applied displacement, from 6 × 10⁻⁴ s⁻¹ to 6 × 10⁻¹ s⁻¹ using an MTS Insight 5, from 8 × 10⁻² s⁻¹ to 8 s⁻¹ using a TA Instruments ElectroForce 3200, and from 6 × 10⁻¹ s⁻¹ to 6 × 10² s⁻¹ using an Instron CEAST 9350 (Figure 1c and Movie S1, Supporting Information). The energy absorption density was calculated as the area under the force–displacement curve normalized by the beam volume (Figure 2, Sections S3 and S4, Supporting Information). The energy absorption density of the polydomain LCE structure increased with strain rate.
according to a power-law relationship, where $\alpha = 0.32$, and also increased 10 times when the displacement rate increased from $10^{-2}$ to $10^1$ mm s$^{-1}$ (effective strain rate: $6 \times 10^{-4}$ to $6 \times 10^{-1}$ s$^{-1}$) (Figure 2b,c). In comparison, the energy absorption density of the same unit cell structure fabricated using PDMS elastomer beams did not vary with strain rate (Figure 2b). Besides, for the highest strain rate in the MTS experiment, the energy absorption density of the polydomain unit cell was nearly an order of magnitude higher than that of the PDMS unit cell (Figure 2b). Note that the LCE structure exhibits pseudo-bistability due to the viscoelastic behavior of the material.$^{[22,23]}$ This means that the structure maintains the snap-buckled configuration for a period then recovers the initial shape. The energy absorption density of all the three types of LCEs increased by two orders of magnitude when the effective strain rate increased from $6 \times 10^{-4}$ to $6 \times 10^2$ s$^{-1}$ (Figure 2c–e).

To understand the rate-dependence of the energy absorption densities of unit cell structures, we characterized the master curves of the storage and loss moduli of the LCE materials with different mesogen alignments using the time-temperature superposition principle (Figure 3a and Figure S9, Supporting Information).$^{[24]}$ The variation in the power-law exponents for the rate-dependence of the energy absorption densities can be explained by the variation in the frequency dependence of the power-law relationship with the strain rate (Figure 2c–e, and Table 1). Beams fabricated from monodomains generally absorbed more energy than polydomain at relatively low strain rates. For the low strain rates of the MTS tests (Figure 2c), the polydomain and 90%∥ beams had similar power-law exponents, while the 90%⊥ beam had a substantially lower exponent. At the moderate strain rates of the ElectroForce tests (Figure 2d), the power-law exponent increased from polydomain through 90%∥ to 90%⊥. In contrast, at the high strain rates of the Instron drop tower tests (Figure 2e), the energy absorption densities of all three different mesogen alignments were less sensitive to the strain rates, compared to the lower strain rate cases.

Figure 1. Mechanical energy absorption via bistable architected LCEs. a) LCEs are synthesized by a Michael addition reaction between diacylate mesogen (RM257), dithiol spacer (C3), and tetrathiol crosslinker (PETMP). A non-equimolar excess of 2 mol% acrylate groups is used to fix mesogen arrangement by additional photo-crosslinking. b) Different mesogen arrangements in the bistable LCE beam are considered by applying different mechanical strains (0% and 90%). The effect of the direction of mesogen arrangement according to the mechanical strain is also investigated by inserting mesogen-arranged LCEs perpendicular (⊥) or parallel (∥) to the beam direction. c) The energy-absorbing capability of LCE structures is characterized over a wide range of strain rates. Three different mechanical testing systems are introduced for an understanding of their strain-rate dependent energy absorption behaviors from quasi-static conditions ($10^{-4}$ to $10^{-1}$ s$^{-1}$) to dynamic conditions up to near $10^3$ s$^{-1}$.
storage and loss moduli. The strain rate in compression tests is proportional to the frequency in dynamic mechanical analysis (DMA). At a few frequencies lower than $10^{-2}$ Hz, the storage modulus of the 90%∥ was larger than for the 90%⊥, and the power-law exponent abruptly decreased to 0.32 for the polydomain and to 0.14 for 90%⊥, respectively. Likewise, at the lower strain rate range ($6 \times 10^{-3}$ to $6 \times 10^{-1}$ s$^{-1}$), the energy absorption density of the 90%∥ beam was larger than that of the 90%⊥. The energy absorption density of the 90%⊥ beams also had the flattest rate dependence in the low strain rate regime (Figure 2c). The storage and loss moduli for all materials showed little variation for high frequencies ($> 10^5$ Hz), indicating glassy behavior. This may explain why the energy absorption densities of the LCE bistable beams were less sensitive to strain rates in the high-rate experiments (Figure 2e). Below $10^3$ Hz (Figure 3a), the storage modulus was larger for the monodomain than polydomain beams. The larger stiffness translated into greater energy absorption density in these comparisons (Figure 2c,d). For frequencies greater than 1 Hz but below $10^3$ Hz, the storage moduli for the polydomain and 90%⊥ increased with increasing frequency with a power-law exponent of about 0.5 (Figure 3a), which is characteristic of the Rouse model. In contrast, the storage modulus for the 90%∥ had a constant power-law exponent of 0.32. The mesogen alignment along the loading direction may inhibit the chain dynamics leading to constant frequency dependence. Since deformation parallel to the director does not induce mesogen rotation, we speculate that the change in the frequency dependence of the modulus observed in the polydomain and 90%⊥ but not in the 90%∥ was caused by a change in dissipation mechanism from viscous chain dynamics to viscous mesogen rotation. Previous works showed that the nematic director relaxation time is longer than the characteristic Rouse time. Therefore, viscous mesogen rotation is likely responsible for the smaller power-law exponent in the frequency dependence of the storage modulus at a lower frequency (Figure 3a) and in the rate-dependence of the energy absorption density at lower rates for the 90%⊥ beam.

Table 1. Power-law exponent values of LCE structures with different mesogen alignments.

| System              | Polydomain | 90%⊥ | 90%∥ |
|---------------------|------------|------|------|
| MTS Insight 5       | n          | 0.323| 0.217| 0.336|
|                     | $R^2$      | 0.983| 0.992| 0.900|
| TA ElectroForce 3200| N          | 0.261| 0.312| 0.348|
|                     | $R^2$      | 0.992| 0.996| 0.982|
| Instron CEAST 9350  | N          | 0.316| 0.357| 0.117|
|                     | $R^2$      | 0.920| 0.911| 0.984|

Figure 2. Mechanical energy absorption for a unit cell. a) Force–displacement curves for architected polydomain LCEs at multiple displacement rates. The area underneath a force–displacement curve indicates energy absorption. The shaded regions indicate the maximum and minimum of 3 specimens. The solid line indicates the average value. b) Energy absorption density of architected polydomain LCE versus PDMS. The energy absorption density follows the power-law relation (represented in red line) for strain rate ($\varepsilon \propto \dot{\varepsilon}^{\frac{3.323}{32}}$, $R^2 = 0.983$). The identical structure composed of PDMS was tested for comparison. c–e) Energy absorption densities of architected LCEs consisting of differently arranged liquid crystal molecules. The energy absorption was characterized by using MTS Insight 5 (c), TA ElectroForce 3200 (d), and Instron CEAST 9350 (e) systems. The lines are the power-law fit between energy absorption density and strain rate.
The effect of mesogen alignment on the molecular relaxation mechanism is summarized schematically in Figure 3b. The relaxation spectra were determined for the different materials from the master curve of the storage moduli (Figure S10, Supporting Information) and applied to finite element simulations of the rate-dependent compression experiments (Figure 3c, Sections S6–S8, Supporting Information). A finite deformation viscoelastic model \cite{28} with a discrete relaxation spectrum was used to describe the behavior of the LCEs. The contributions to energy absorption from the stored energy and the viscous dissipation were computed for the viscoelastic model (Section S7, Supporting Information). Both the stored and dissipated energy densities increased with strain rate for all materials for the strain rates $8.3 \times 10^{-4}$ to $8.3 \times 10^{2}$ s$^{-1}$ in experiments. Both the stored and dissipated energy densities of the polydomain are smaller than those of 90%∥ and 90%⊥ over the entire range of strain rates, which is consistent with the frequency dependence of the master curve of the storage modulus of the three types of LCEs (Figure 3a and Figure S9, Supporting Information). During the low-rate regime corresponding to MTS test in Figure 2c, the stored energy density for all three materials increased less than one order of magnitude, whereas the dissipated energy density increased more than an order of magnitude. Compared to stored energy, viscous dissipation was more sensitive to strain rate during the low strain rate range, regardless of mesogen alignment (Figure 3c). In contrast, as strain rate increased, the rate-dependence of viscous dissipation for 90%⊥ became flatter, but that of 90%∥ did not change. It is likely because in the 90%⊥ case, there is also a mesogen rotation dissipation mechanism, whereas in 90%∥, there is only viscous chain dynamics. Due to the increasing contribution of stored and dissipated energy (Figure 3c), the total energy absorption increased with strain rates (Figure 2c–e).

We next investigated the energy absorption of multilayered bistable structures by arranging the unit cells with polydomain LCE beams in $2 \times n$ arrays, where $n$ is the stacking number (Section S2, Supporting Information). Figure 4a shows a series of snapshots of a $2 \times 2$ structure under uniaxial compression at an effective strain rate of $2.38 \times 10^{2}$ s$^{-1}$ (Movie S2, Supporting Information). Both layers were initially compressed together. However, instead of both layers buckling simultaneously, the top layer began to buckle whereas the second layer recovered (i.e., straightened). As the top layer collapsed, the bottom layer again compressed and buckled. The sequential buckling of the layers is also evident in the plot of the force-displacement curves (Figure 4b), which shows two distinct peaks. The larger first peak corresponds to the initial compression of the two
layers and buckling of the top layer, while the second smaller peak marks the buckling of the bottom layer. We also observed a nonuniform buckling behavior at the same strain rate for the taller $2 \times 3$ and $2 \times 4$ structures. The force–displacement curves had 3 and 4 peaks for the $2 \times 3$ and $2 \times 4$ structures, respectively (Figures S7 and S8, Supporting Information). The nonuniform buckling of the layers caused the energy absorption density to increase with the stacking number $n$ (Figure 4c). This effect was not observed for the multilayered structure with elastomeric PDMS beams,[1] where the energy absorption density was constant regardless of stacking number and strain rate, and total energy absorption scaled linearly with stacking number. The energy absorption density of stacked LCE structures also followed a power-law relationship with the strain rate (Table 2).

To better understand the mechanisms underlying the increase in the energy absorption density with the stacking number, we explored two distinct approaches: (1) varying the number of stacked layers and (2) modifying the gradient in beam thickness. These strategies enabled us to systematically investigate the impact of structure geometry on energy absorption performance.

**Figures 4a-4f**

- **Figure 4a**: Experimental snapshots of a $2 \times 2$ stacked LCE structure under compressive loading at a strain rate of $2.38 \times 10^{-2}$ s$^{-1}$: (i) and (ii) initially both layers collapsed simultaneously, (iii) the bottom layer slightly recovered back (i.e., straightened) while the top layer kept collapsing, (iv) both layers collapsed till fully deformed.
- **Figure 4b**: The experimental force–displacement curves of $2 \times 2$ stacked LCE structures at different strain rates. The shaded region indicates the range of experimental measurement for at least three specimens. The solid lines indicate the average values.
- **Figure 4c**: The energy absorption density of stacked LCE structures plotted as a function of strain rates for different stacking numbers from $2 \times 1$ to $2 \times 4$ (experiment).
- **Figure 4d**: The total energy absorption density, dissipated energy density, and stored energy density of stacked LCE structure plotted as a function of stacking number at a strain rate of $2.27 \times 10^{-2}$ s$^{-1}$ (simulation).
- **Figure 4e**: The force–displacement curves of $2 \times 8$ stacked LCE structures with uniform and graded beam thicknesses for a strain rate of $2.27$ s$^{-1}$ (simulation).
- **Figure 4f**: The difference in the energy absorption density between graded and uniform cases for $2 \times 2$, $2 \times 4$, and $2 \times 8$ stacked LCE structures plotted as a function of strain rates (simulation). The horizontal dash line passing through zero indicates the case where the energy absorption density of the structures with graded beam thickness is equal to those with uniform beam thickness.
number, we simulated the compression tests and computed the rate-dependence of the stored energy density and the dissipated energy density for different stacking numbers (Sections S7 and S8, Supporting Information). As the stacking number increased, the dissipated energy density increased, while the stored energy density remained constant (Figure 4d and Figure S19, Supporting Information). As in experiments, the viscoelastic behavior of LCE introduced non-uniformity in the stress-state of the beams to produce non-uniform buckling. The buckling of some layers caused other layers to recover (i.e., straighten) then buckle again after the collapse of the preceding layers (Movies S2 and S3, Supporting Information). This loading-unloading cycle increased the viscous dissipation without changing the stored energy, thus enabling increased total energy absorption density. For taller structures with more than 3 layers, the unit cells in adjacent layers again buckled together, resulting in a smoother force–displacement curve, with fewer distinct peaks than the stacking number and the energy absorption density to plateau (Figures S16 and S24, Supporting Information). As a result, the energy absorption density saturated at a stacking number of 3.

To further promote this dissipation mechanism for higher stacking numbers, we varied the thicknesses of the LCE beams from the bottom to the top layers to ensure sequential buckling of the different layers (Movies S3 and S4, Supporting Information). The total volume of the beams was kept the same as for models with uniform beam thickness. Specifically, for a $2 \times 8$ structure the beam thickness of a subsequent layer was 0.05 mm larger than the preceding layer, viewing from top to bottom. The graded structure buckled sequentially from the top to the bottom, producing eight distinct peaks in the force-displacement curve (Figure 4e and Movie S4, Supporting Information). The first and largest peak of the force-displacement curve was caused by the initial simultaneous compression of all 8 layers and the subsequent buckling of the top layer. The 2nd peak arose from the continued collapse of the previous layer and the buckling of the 2nd layer, and so forth. As a result, the peak force exhibited a monotonic increase with the number of layers. Therefore, by making graded structures with varying beam thicknesses, we could ensure sequential buckling of different LCE layers to enhance the dissipated energy density and thus the total energy absorption density (Figure S25, Supporting Information). The uniform $2 \times 8$ stacked structure, by contrast, deformed simultaneously resulting in the first large peak and there is no match between the number of layers and the number of peaks as shown in Figure 4e. Note that the energy absorption density did not saturate at the stacking number 3 at the highest strain rate of the experiment (Figure 4c), which was most likely due to the sample fabrication issue as there were uncontrollable variations of the beam dimensions and their mechanical properties of the different layers. Specifically, the experimental samples did not have the same beam thickness across the different layers (see Table S1, Supporting Information). This effect is consistent with the findings of the FEM simulations that showed grading the beam thickness ensures that the energy absorption density can increase unbounded with the stacking number.

To quantitatively compare the graded and uniform structures, we calculated the difference in the energy absorption density between graded and uniform cases at different strain rates (Figure 4f). The difference in the energy absorption density increased exponentially with strain rates. At the highest strain rate in Figure 4f, the energy absorption density of the $2 \times 8$ graded structure was 45% higher than that of the $2 \times 8$ uniform structure. Furthermore, the energy absorption density of graded structures did not plateau at higher stacking numbers (Figure S26, Supporting Information) This synergistic interaction between viscoelastic dissipation and snap buckling provides new opportunities for the design of the material architecture to enhance energy absorption.

### Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

| Stack number | $a$ | $b$ | $R$ | $2$ |
|--------------|-----|-----|-----|-----|
| 1            | $0.63 \times 10^1$ | $0.198$ | $0.925$ |
| 2            | $1.04 \times 10^1$ | $0.264$ | $0.999$ |
| 3            | $1.10 \times 10^1$ | $0.231$ | $0.982$ |
| 4            | $1.37 \times 10^1$ | $0.249$ | $0.972$ |
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Conflict of Interest
The authors declare no conflict of interest.

Data Availability Statement
The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords
energy absorption, liquid crystal elastomers, metamaterial, power-law relation, stacking effect, viscoelasticity

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