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Engineering the shape and structure of materials by fractal cut

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In this paper we discuss the transformation of a sheet of material into a wide range of desired shapes and patterns by introducing a set of simple cuts in a multilevel hierarchy with different motifs. Each choice of hierarchical cut motif and cut level allows the material to expand into a unique structure with a unique set of properties. We can reverse-engineer the desired expanded geometries to find the requisite cut pattern to produce it without changing the physical properties of the initial material. The concept was experimentally realized and applied to create an electrode that expands to >800% the original area with only very minor stretching of the underlying material. The generality of our approach greatly expands the design space for materials so that they can be tuned for diverse applications.

expandability | patterning | flexibility | differentiation | deformability

he physical properties of materials are largely determined by structure: atomic/molecular structure, phase distribution, internal defects, nano/microstructure, sample geometry, and electronic structure. Among these, engineering the geometry of the sample can provide a direct, intuitive, and often materialindependent approach to achieve a predetermined set of properties. Metamaterials are fabricated based on geometric concepts (1–16). In two dimensions, periodic geometries have been adopted to tune the mechanical properties of membranes (3–8, 10, 12–14). From simple shapes such as circles (3), triangles (6, 7, 12, 13), and quadralaterals (4, 5, 14) to more complex shapes (8, 10), a broad range of mechanical behavior has been observed, including pattern transformation, negative Poisson's ratio (auxetic), elastic response, and isostaticity. *Origami* and *kirigami*, the arts of paper folding and paper cutting, create beautiful patterns and shapes that have attracted the attention of scientists to two-dimensional materials (e.g., graphene, polymer films, and so on) (11, 17–19). However, application of conventional origami and kirigami approaches to achieve desired material response requires complex cutting and/or folding patterns that are often incompatible with engineering materials. In this paper we propose an advanced approach to the design of two-dimensional structures that can achieve a wide range of desirable programmed shapes and mechanical properties.

This study starts from the question, Can we design twodimensional structures that can be formed by simply cutting a sheet, that can morph into a specific shape? In nature, many biological and natural system (20) can be found that use hierarchical structure to produce different properties and/or shapes. One such example is a stem cell. An embryonic, pluripotent stem cell can differentiate into any type of cell in the body (21). By recursively dividing, the stem cell can transform into particular cell types or remain unspecialized with the potential to transform. For a material, one aspect of recursive hierarchical geometry was recently discussed for applications in flexible electronics (22). Here, by analogy to the stem cell, we demonstrate that starting from a simple sheet of material (the pluripotent state) it is possible to apply different hierarchical cut patterns (differentiation) to achieve a wide range of macroscopic (unipotent) shapes. In particular, we focus on "fractal cut" patterns that allow for precise control of differentiated material structures. Our goal is to broaden the design space for engineered materials and systems for a wide range of applications, such as flexible/stretchable devices, photonic materials, and bioscaffolds. We illustrate the concept through numerical simulation, theory, and experimental realization.

Basic Principle: Rotating Units

For simplicity, we focus on a base material in the form of a flat, flexible sheet and subject it to a prescribed cut pattern. The essence of the design is that cuts divide the material into rotating units, depending on the cut pattern (4, 5), as exemplified in Fig. 1A. The units (e.g., squares or triangles) between cuts are effectively rigid and the connections between these units behave as (nearly) free rotational hinges, such that the deformation of the structure (e.g., biaxial or uniaxial stretching) occurs primarily through rotation of the units, rather than by significant deformation of the units themselves. The final material morphology, determined by the cut pattern, is determined through moment equilibrium (23). There is a maximum stretch ratio (strain) that can be achieved by rotation for any specific cut pattern beyond which the units themselves will deform; this latter stage of deformation is conventional and is not of interest here.

Significance

Most materials can be stretched to a small degree, depending on their elastic limits and failure properties. For most materials the maximum elastic dilatation is very small, implying that the macroscopic shapes to which an elastic body can be deformed is severely limited. The present work addresses the simple modification of any material via hierarchical cut patterns to allow for extremely large strains and shape changes and a large range of macroscopic shapes. This is an important step in the development of shape-programmable materials. We provide the mathematical foundation, simulation results, and experimental demonstrations of the concept of fractal cut. This approach effectively broadens the design space for engineered materials for applications ranging from flexible/stretchable devices and photonic materials to bioscaffolds.

Author contributions: Y.C. and I.-S.C. conceived the concept of the fractal cut and designed research; Y.C., J.-H.S., A.C., T.A.K., S.Y.L., S.Y. and I.-S.C. contributed experimental realization; V.K. and D.J.S. contributed the geometric model; Y.C., H.N.H., and I.-S.C. performed finite element simulations; Y.C., V.K., J.L., I.-S.C., and D.J.S. analyzed data; and Y.C., S.Y., I.-S.C., and D.J.S. wrote the paper.

Conflict of interest statement: Y.C. and I.-S.C. have filed a patent application relating to expandable electrodes based on the fractal cut pattern.

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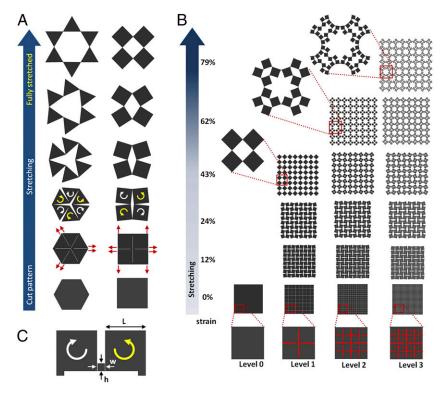


Fig. 1. Basic principles of the cut design. (A) Cuts separate the material into rotating units, with connectivity dependent on the cut pattern (assuming freely rotating point hinges). A hexagon can be divided into six smaller triangles in a pattern that can be repeated to fill space or a square can be divided into four smaller squares. Upon equal-biaxial stretching, each unit rotates clockwise (white arrows) or counterclockwise (yellow arrows), yielding an expansion of the original structure. Expansion continues up to a maximum level by pure unit rotation (minimal strain within the structural units). The structures are fully stretched when moment equilibrium is achieved. If the units are rigid, no further expansion is possible. (B) Finite element calculations for finite size hinges (see C) in silicone rubber. The cuts can be combined in a multilevel hierarchy. The level-0 structure, which has no embedded cuts, is cut into four smaller squares to make the level-structure. Each such unit is cut into four smaller squares to make a higher-level structure, up to an arbitrary degree/level of hierarchy, N. The level-0 structure does not expand because no rotation is possible upon biaxial stressing. In the level-1 structure, each unit can rotate up to 45° . The corresponding lateral strain is 43%. The maximum lateral strains for n = 2 and 3 are 62% and 79%, respectively. These finite element results were confirmed by comparison with the geometric model (rigid units and free hinges, as per A), the macro/microscopic experiments, and analytical predictions (Supporting Information). The pore shape, pore size, and apparent density vary with hierarchy level. (C) A schematic of the finite hinge geometry used in the finite element method calculations (B) and in the experiments.

Fractal Cut Pattern: Hierarchy

In this paper we discuss two classes of cut patterns: hierarchical and motif alternation. The hierarchical pattern concept is illustrated in Fig. 1B for a simple pattern of cuts producing square units. Such square units can be subdivided into smaller squares by repeating the cut pattern within the original square. Although the subdivision can, in principle, go on ad infinitum, creating a true fractal cut pattern (20), we focus on patterns of finite hierarchy degree or level (i.e., the number of times the same cut pattern is reproduced on the units left by the preceding cuts). Increasing the hierarchy level leads to increasingly complex structures and increased expandability. Along with expandability, pore shape, apparent density, and elastic stiffness (assuming the hinges have some resistance to rotation) all vary with the hierarchy level. Examination of Fig. 1B shows that the expansion at one level is largely exhausted before the expansion at the next level of the hierarchy operates (cf. level-1, -2 and -3 structures at a biaxial strain of 0.24). (Note: we focus on square units in two dimensions in this discussion, but application to triangular units in two dimensions and cubical units in three dimensions are shown in Supporting Information.)

The variables that determine the final structure of the stretched sheet are the rotation angles between rotating units. The number of independent variables increases with increasing level. The level-0 structure, which has no cuts has 0 independent variables (i.e., no degrees of freedom $F_0 = 0$). The level-1 structure has

one degree of freedom (one independent angle determining the rotation of all units), $F_1 = 1$. The level-2 and -3 square structures have two and six degrees of freedom, $F_2 = 2$ and $F_3 = 6$, respectively. The number of degrees of freedom grows as $F_N = 4F_{N-1} - 2$ or $F_N = (4^{N-1} + 2)/3$ (for n > 1), where N is the hierarchy level. This implies that for any strain smaller than some maximum the structure with free hinges is not fully determined (i.e., there are multiple sets of angles that can lead to exactly the same strain) for any $n \ge 2$ (see *Supporting Information* for a specific example).

Whereas "free hinges" is an idealization, in any real material application the hinges have finite rotational stiffness. Consider the case where the "hinges" consist of the incompletely cut units, as illustrated in Fig. 1C. The rotational (bending) stiffness of the hinge is proportional to h^3 (Fig. 1C). In any such real case the structure is fully determined at any strain. The maximum stress in the hinge during rotation (bending of the ligament) is proportional to h/w. Hence, appropriate hinge design represents a compromise between hinge failure and hinge stiffness. The geometric parameters describing the hinges in the finite element method calculations and experiments are reported in *Supporting Information*. We note that the design must also be sensitive to the actual choice of materials, in particular the stiffness and the fracture and yield strengths. Although elastomeric systems are obvious choices for such applications, they could also be

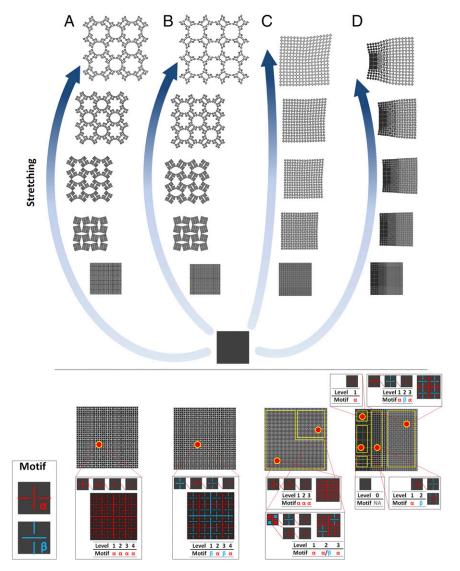


Fig. 2. Two cut motifs in a hierarchical structure. The cuts shown in red (blue) refer to the α -motif (β -motif). The β -motif is a 90° rotation of the α -motif in two dimensions. (A) The pure α -motif has a maximum lateral strain of 108% at level 4, $\varepsilon_4(\alpha\alpha\alpha\alpha)$. (B) The alternating α - and β -motif at level 4, $\varepsilon_4(\beta\alpha\beta\alpha)$, yields a maximum lateral strain of 130%. (C) Inhomogeneous deformation can be realized by varying the α - and β -motif in a level-3 structure or (D) by mixing regions of different cut levels as indicated at the bottom of the figure. These examples were determined from finite element calculations with material parameters for silicone rubber.

fabricated from metals provided h/w is sufficiently small to limit the stresses in the hinge to be below the yield strength.

Fractal Cut Pattern: Motif

Besides hierarchy, another design parameter is the cut motif. In the previous section, the cut motif was constant (square or triangular), as indicated by the red lines in Fig. 1B (the α -motif). This same motif applied homogeneously leads to the same unit rotation pattern across the entire structure (white and yellow arrows in Fig. 1A). The number of degrees of freedom F grows monotonically with the hierarchy level N, whereas the increment of rotation angle becomes smaller. For example, the rotation angle of the smallest unit in the level-1 structure in Fig.1B is 45°, but the rotation angle for the smallest unit in the level-2 structure is ~27°, ~12° for the smallest unit in the level-3 structure, and ~8° for the level-4 structure in Fig. 2A (see Supporting Information for details). This implies a finite limit to the expandability of structures with identical motifs.

Another motif, β , can be formed by rotating the α -motif by 90°, as shown in blue at the bottom of Fig. 2. In this motif the square units rotate in opposite directions relative to those in the α -motif. The combination of α - and β -motif between levels, hence, produces alternating rotation directions of the units, leading to larger rotation angles and strains at higher levels. We denote the strain at each level i in an N-level structure as $\varepsilon_N(x_1x_2...x_N)$, where x_i denotes the motif (e.g., x_i refers to the α - or β -motif). For example, the maximum lateral strain in the level-4 structure consisting of a single cut motif is $\varepsilon_4(\alpha\alpha\alpha\alpha) = \varepsilon_4(\beta\beta\beta\beta) = 108\%$ (Fig. 2A), whereas for the alternating motif it is $\varepsilon_4(\beta\alpha\beta\alpha) = 130\%$ (Fig. 2B).

Engineering Shape and Structure via Fractal Cut

Hierarchical levels and motifs provide the basic palette that can be used to draw (i.e., cut pattern) on a blank canvas (or material sheet). Different motifs and levels give different rotation patterns and strains, allowing for tunability. For the case of two motifs, we can evaluate the total number of ways

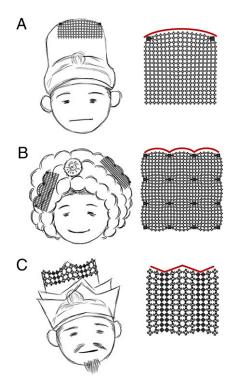


Fig. 3. Fractal cut Korean traditional hat and hairstyle realized by engineering the distribution of hierarchy and motif. (A) A Tangon hat from combination of $2 \cdot \epsilon_0 + 68 \cdot \epsilon_3(\alpha \beta \alpha) + 11 \cdot \epsilon_4(\beta \alpha \beta \alpha)$ units, (B) a Gache hairstyle from a combination of $16 \cdot \epsilon_0 + 132 \cdot \epsilon_3(\alpha \beta \alpha) + 21 \cdot \epsilon_4(\beta \alpha \beta \alpha)$, and (C) a Jeongjaguan hat from a combination of $12 \cdot \epsilon_1(\alpha) + 24 \cdot \epsilon_4(\beta \alpha \beta \alpha)$ as determined from finite element calculations with material parameters for silicone rubber.

to cut the material. At the first level, where the α - or β -motif can be applied, there are only two permutations, i.e., $V_1=2$. At level 2, each of the four subsquares has one of two motifs

(i.e., 2^4 possibilities). Therefore, in a level-2 structure, $V_2 = 2^4 \times 2 = 32$. More generally, a level N structure with two motifs has

$$V_N = 2^{\sum_{n=1}^{N} 4^{n-1}} = \prod_{n=1}^{N} 2^{4^{n-1}} = 2^{\frac{4^{N-1}}{3}}.$$
 [1]

Here, level and motif distributions represent a mechanism for pluripotency. The original sheet (intact square) is pluripotent; when the fractal cut design is embedded, the sheet becomes unipotent. Upon stretching, the rotation of the units activates the differentiation. The final sheet shape can be programmed. For example, Fig. 2C shows the nonuniform expansion of a level-3 structure with an inhomogeneous combination of α - and β -motifs, and Fig. 2D shows the expanded shape resulting from a mixture of different hierarchy levels and motifs. We can exploit the pluripotency of a single square sheet to reproduce shapes of considerable complexity. Fig. 3 applies such an approach to reproduce traditional Korean hats and hairstyle.

Experimental Realizations

Structural differentiation was experimentally realized as shown in Fig. 4A–D. We fabricated square sheets of silicone rubber with four different fractal cut patterns using three-dimensional printed molds. Fig. 4A–D correspond to the simulated patterns from Fig. 2A–D. By stretching, the square sheets show final shapes that very nearly match the simulation results. Obviously, the concept of fractal cut is not confined to a specific material system or to a specific feature size. For example, reducing the smallest feature scale in the level-4 structures in Fig. 4A and B from 2 mm to 40 μ m using photolithography to make molds into which polydimethylsiloxane (PDMS) sheets were cast leads to identical differentiation (see Fig. 5 and the *Supporting Information* for more experimental details). Hence, the present approach to forming highly expandable pluripotent materials can be applied on the macro- or microscale.

Discussion

Our pluripotent material approach provides an effective means for the design of structural platforms for stretchable and flexible devices.

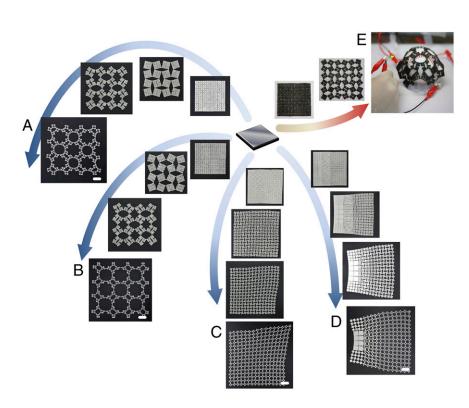


Fig. 4. Macroscopic experimental realization. Silicone rubber sheets differentiated with patterned cuts identical to those used in the finite element calculations. The smallest feature size (hinge width) is 2 mm. A–D correspond to the cut patterns in Fig. 2 A–D (these samples were pinned open to show expanded states). The correspondence between simulation and experiment is excellent. (E) A proof of concept for a stretchable electrode platform is shown by embedding $\varepsilon_3(\alpha\alpha\alpha)$ fractal cuts in a silicone rubber sheet coated with a thin conducting film and conformably wrapping the structure around a spherical baseball without wrinkles. The green LED light remains lit after wrapping. (The white lines in the expanded structure images are 40 mm.)

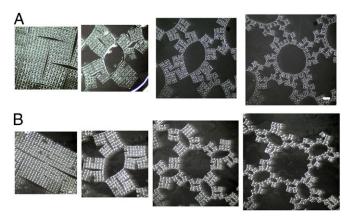


Fig. 5. Microscopic experimental realization. PDMS sheets differentiated with patterned cuts. The smallest feature size (hinge width) is 40 µm. (A) One unit of the homogeneous $\varepsilon_4(\alpha\alpha\alpha\alpha)$. (B) One unit of the $\varepsilon_4(\beta\alpha\beta\alpha)$. The correspondence between finite element simulation, macroscopic experiment, and microscopic experiment is excellent (cf. Figs. 2, 4, and 5). (The white line in the expanded structure image is 200 µm.)

Because stretching occurs by unit rotation rather than deformation, the material in the structure is inherently (nearly) strain-free (except at the hinge points); this is essential for stretchable platforms. It can also be strained without buckling. Thus, deformation of the structure will not alter the physical properties/function of the materials deposited on top of the units. Fig. 4E shows a proof of concept of a stretchable electrode with a fractal cut. We deposited a conductive film of multiwall carbon nanotubes on a silicone rubber sheet with an embedded homogeneous n = 3, α -motif. A light-emitting diode (LED) continues to be powered through the conductive film as the cut silicone rubber sheet is stretched over a spherical baseball (see Supporting Information for more experimental details). The conformal wrapping of the sheet around a nonzero Gauss curvature object (a sphere in this case) leads to nonuniform stretching (and nonuniform opening patterns), which can easily be accommodated by the fractal cut sheet (an example for other, nonbiaxial loads is shown in Supporting Information). Our approach to stretchable/flexible substrates differs from others in the literature, where expansion and conformal wrapping of a flexible device consisting of rigid components connected through stretchable elements (e.g., springs and serpentines) (15, 24, 25). In our systems, because the deformation is based on unit rotation we can fabricate a highly expandable device (e.g., ~800% areal expansion from the level-6 (βαβαβα) structure; see *Supporting Information* for details) by placing conventional hard devices (e.g., battery, circuit, LED, etc.) on the rotating units without sacrificing device performance during large deformation.

of rigid units meeting at free hinges, this is only an idealization.

Although an ideal fractal cut material expands by the rotation nology and Future Planning Grant 2013008806.

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Our experimental realizations, however, are made with cuts that leave a finite ligament between the units. This has two consequences: First, these ligaments are strained and, second, this provides a small resistance to rotation (i.e., the hinges are not completely free). Nonetheless, a comparison of the experimental realization, its finite element simulation, and the rigid-unit/freehinge model are in excellent correspondence (see Supporting Information for details). This implies that the theoretical idealization is not unreasonable and the approach can be applied to any material where hinge-like structures are possible; here, for simplicity the concept was demonstrated with silicone rubber and PDMS. Material design to achieve target expandability distributions/morphologies is an inverse problem in cut geometry. Unlike many materials design problems, the inverse problem for fractal cut structures is relatively straightforward with the simple design palette (cuts) described here and the straightforward calculations implied by the theoretical idealization.

Although the present results focused on two-dimensional sheets with square-based units as a starting point, the same approaches can be applied with (i) a different two-dimensional base unit [see Supporting Information for a triangular (kagome) lattice example] and (ii) three-dimensional materials using one of the many recent technological advances in three-dimensional printing (see Supporting Information for a free-hinge numerical example based on rotating cubes). By prescribing the geometry of cuts in a sheet we can simply control not only the meso/nano structure of a sheet but also engineer all of the properties that map to its structure, including those associated with shape (pore size, pore shape, macrogeometry, and maximum strain), mechanical properties (full stiffness tensor), and even material properties coupled with structures (electrical, photonic, and acoustic properties). Many of these require additional manipulation of the connections between the rotating units (e.g., stiffness depends on finite length of the material in hinges). Designing actuation or prerotations into the structure can further enhance the flexibility and functionality of cut structures for various applications.

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