Materials with structural hierarchy

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Many natural and man-made materials exhibit structure on more than one length scale; in some materials, the structural elements themselves have structure. This structural hierarchy can play a large part in determining the bulk material properties. Understanding the effects of hierarchical structure can guide the synthesis of new materials with physical properties that are tailored for specific applications.

HIERARCHICAL solids contain structural elements which themselves have structure. The hierarchical order of a structure or a material may be defined as the number (n) of levels of scale with recognized structure. For $n=0$, the material is viewed as a continuum for the purpose of analysis of physical properties; $n=1$ (first-order) could represent a latticework of continuous ribs or the atomic lattice of a crystal. Hierarchical structure can arise in natural and in man-made materials. In the latter the structural hierarchy may be intentional or unintentional. The simplest conceptualization of hierarchical structure is descriptive: to recognize that structural features occur on different size scales. At the next level of sophistication, the idea of hierarchical structure can be used in analysis to determine physical properties of the material or the structure. At each level of the structural hierarchy, one may model the material as a continuum for the purpose of analysis, although strictly speaking such an assumption is warranted only if the structure size at each level of the hierarchy is very different. Finally, the idea of hierarchical structure can be the basis for synthesizing new microstructures which give rise to enhanced or useful physical properties. Benefits can include improved strength and toughness, or unusual physical properties such as a negative Poisson's ratio. These structures are considered to be fractal-like[1], but they are not true fractals as n remains finite and the solid volume fraction does not go to zero even for large n.

The idea of macroscopic hierarchical frameworks can be traced back at least to Eiffel's design for his tower[2] (Fig. 1) and to bridges such as the Garabit viaduct. The Eiffel tower is third-order, and has a relative density ρ/ρ_0 (density ρ as mass per unit volume of the structure divided by density ρ_0 of material of which it is made) just 1.2×10^{-3} times that of iron[2] (which is weaker than structural steel). The rationale for the use of small girders in such a large structure was attributed to ease of construction[3], although it had also been suggested by Mandelbrot[1] that Eiffel perceived a structural advantage. For comparison, the World Trade Center (New York) and the Pompidou Centre (Paris), both first-order, contain a volume fraction of structural steel[4] $\rho/\rho_0=5.7\times10^{-3}$. The World Trade Center contains steel with a yield strain ε_y of 0.0033, 2-3 times as strong as 'mild' structural steel.

A more recent example is a proposal by Dyson[5] to construct hierarchical frameworks in outer space. Dyson presented scaling arguments to the effect that very large structures could be constructed with low mass. Stress analysis of elastic buckling in hierarchical truss structures was to come later[6,7]. In modern structural engineering, however, the tendency seems to be away from hierarchical structures; although these contain less material to achieve a desired strength, the costs associated with fabrication and maintenance currently exceed any saving in material cost.

Dense hierarchical materials

Composites and polycrystals. Practical fibrous composites commonly have a low order of hierarchical structure in which fibres are embedded in a matrix to form an anisotropic sheet or lamina; such laminae are bonded together to form a laminate (Fig. 2a). In the analysis of fibrous[8-10] composites, the fibres and matrix are regarded as continuous media when one is analysing the lamina; the laminae are then regarded as continuous in the analysis of the laminate. The stacking sequence of laminae and the orientation of fibres within them governs the anisotropy of the composite. A similar continuum assumption is used in the analysis of particulate composites[11] and of foams[12]. Inorganic crystalline materials have a 'hierarchy' of structural features[13] such as grain boundaries between crystals of sizes ranging from millimetres down to micrometres, dislocations, and point defects such as vacancies on the atomic scale. These structural features give rise to viscoelastic behaviour[14,15] manifested as attenuation of stress waves or damping of vibration at different frequencies. Polycrystalline materials can now be synthesized with a distribution of grain sizes less than 1 μm (nanocrystalline materials)[16-19]. The small grain size, and hence large interface area, gives rise to desirable properties such as superplasticity (in which large irreversible deformation can occur without fracture), and improved strength and toughness. Small grain size also implies short diffusion distances, allowing processes that depend on diffusion, such as sintering, to occur at lower temperatures than would otherwise be possible.

Hierarchical laminate structures have been analysed theoretically to approximate the stiffness of polycrystalline aggregates[20], and for exploring bounds on the electrical conductivity of poly-

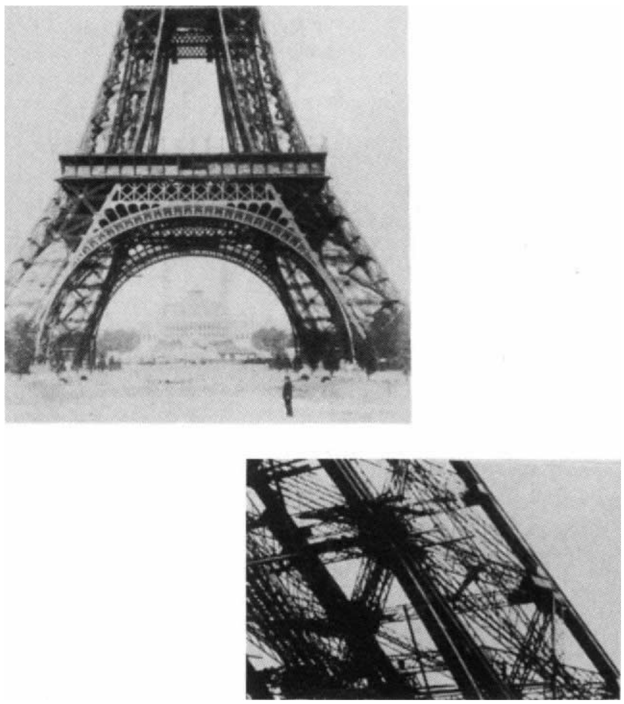

FIG. 1 Hierarchical structure in the Eiffel tower, after Loyrette[2]. Inset: detail of leg.

crystals[21,22] and the elastic stiffness of polycrystals[23] and composites[24]. In these laminates, each lamina is composed of further laminae (Fig. 2*b*). For elastically isotropic hierarchical laminates, it is possible to attain[24] the theoretical upper or lower bounds[25] on the stiffness. These laminates are considered to be a mathematical tool rather than practical composites because widely differing length scales must be chosen to justify the assumption that each level is a continuum[23]. Negative Poisson's ratios, which imply that the material becomes fatter in cross-section when stretched, are predicted in hierarchical laminates[26] with a chevron structure (Fig. 3*a*). The physical mechanism for the unusual Poisson effect is illustrated by the hinged framework which unfolds under tension (Fig. 3*b*). One can achieve, with these laminates, Poisson's ratio values approaching the lower limit of -1 for mechanically isotropic materials.

Polymers. Polymers can exhibit structural hierarchy on the molecular, ultrastructural and microstructural levels[27]. In crystalline polymers, there are spherulites on the scale of tens of micrometres, the spherulites themselves contain a lamellar texture and the molecules within the lamellae contain structure. Amorphous polymers have structure on the molecular scale only[27]. When they are irreversibly deformed, however, crazing occurs and the process can be understood with the aid of a hierarchical approach which can deal with the multiple size scales involved. Crazes are bridged by nanoscale microfibrils whose properties are important. At the macroscale the crazed material can be considered as a composite. In covalent amorphous solids, the concept of hierarchical order has been used to aid the classification of order[28] into short-range (2–5 Å), medium-range (5–20 Å) and long-range ($\geq$20 Å).

Biological materials. Human compact bone is a natural composite which exhibits a rich hierarchical structure[29,30] (Fig. 4). On the microstructural level are the osteons[31], which are large (200 μm diameter) hollow fibres composed of concentric lamellae and of pores. The lamellae are built of fibres, and the fibres contain fibrils. At the ultrastructural level (nanoscale) the fibres are a composite of the mineral hydroxyapatite and the protein collagen. These specific structural features have been associated with various physical properties. For example, the stiffness[32] of bone arises from the composite structure of mineral microcrystals and protein (principally collagen) fibres. Slow creep[33] results from slip at cement lines between osteons. The cement lines as weak interfaces impart a degree of toughness[34] to bone. As for pores, the lacunae are ellipsoidal pores which provide space for the osteocytes, the living cells of bone. The bone cells at this level of scale permit bone tissue to remodel its structure in response to prevailing stresses[30]. Haversian canals are cylindrical pores containing blood vessels which nourish the tissue. Canaliculi are fine channels radiating from the lacunae. Mechanical stress due to physical activity is considered to be important in pumping nutrients through these channels[35]. The pore structure of bone is essential in maintaining its viability and consequently its ability to adapt to mechanical stress. A two-level hierarchical analytical model[36] has been used to predict its anisotropic elasticity; it successfully modelled how bone stiffness depends on the orientation of applied stress with respect to the osteon axis.

Other examples of natural hierarchical materials include wood[37,38], tendon[27], trabecular (spongy) bone and bamboo. Of these, only tendon may be regarded as 'dense'; the others are cellular. Tendon consists of collagen which on a molecular scale is similar to that of bone[27]. The triple helical collagen macromolecule is formed as a result of the amino acid glycine occupying every third unit. The strongest intermolecular attractions occur when neighbouring molecules are shifted by 67 nm, the 'stagger' which is responsible for the banded appearance of collagen observed by electron microscopy. Assembly of subfibrils into fibrils is thought to be controlled at least in part by the primary structure of collagen. In tendon, the collagen forms fibres which are organized into mostly parallel fibre bundles of progressively larger size. The larger-scale organization is attributed to interaction with noncollagenous components such as proteoglycan matrix. The fibres are not perfectly aligned; they form a wavy or crimped structure which confers on the tendon an initial compliance as the fibres straighten under load. The damage processes that governs the strength and toughness of tendon involve structural elements over the full hierarchical range of sizes.

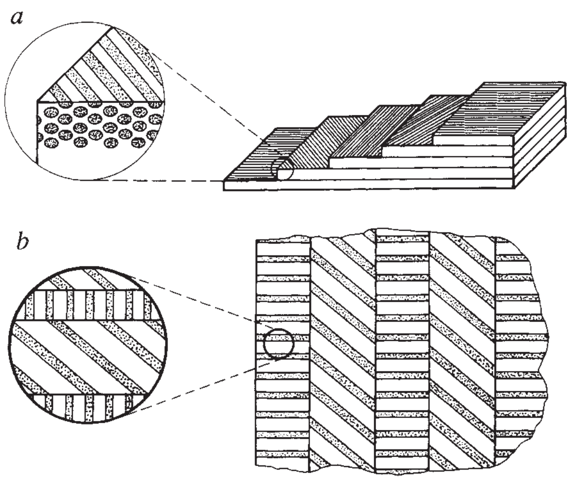

FIG. 2 *a*, Hierarchical structure of order two in a practical fibrous laminate. Parallel fibres form a lamina; laminae of different orientations are stacked to form a laminate with specified anisotropy. *b*, Hierarchical laminate, adapted from Milton[23]; order three shown. Each lamina contains a sublaminate. These laminates are used in mathematical demonstrations of attainable material properties in composities.

Role of the largest structural elements

Structure may be present on many size scales, but the largest structural elements often have a unique role. If the largest structure is not negligible in size compared with the object itself or a crack or hole in the object, the classical continuum view may no longer describe the situation adequately. When deformed elastically, objects with large structural elements may exhibit size effects in bending and torsion[39]: 'classically', the rigidity of rods in bending or torsion should be proportional to the fourth power of the diameter, but slender rods can be stiffer than this. Moreover the stress concentration predicted in classical elastic solids near holes and notches is alleviated in some materials with microstructure[39]. This is beneficial in structural materials. In some foams, incomplete cells near a cut surface contribute to the volume but not to the stiffness or strength[40] so that small objects are less stiff than expected from classical continuum analysis (the converse of the slender rods). In hierarchical composites, the largest structural elements such as fibres[41,42] or particulate heterogeneities[43] seem to govern the fracture toughness and the localization of microdamage. Classical elasticity theory has no length scale associated with it. More general continuum theories such as Cosserat (micropolar) elasticity[44,45] allow rotation of points in the continuum as well as translation, and contain characteristic lengths as well as stiffnesses among the material constants. Physically the additional freedom in the continuum corresponds to twisting or bending motions in the fibres or ribs in the material microstructure. Generalized continuum theories offer predictive power in dealing with nonclassical phenomena[39-43], and may be of use in future analysis of hierarchical materials in which one relaxes the assumption that the structure size at each hierarchical level is very different.

Hierarchical cellular materials

Cellular solids. Cellular solids are composites in which one phase is solid and the other is empty space, or possibly a fluid. Natural examples are rocks, wood and bone. Porous rocks can have a wide range of pore sizes[46], but the structure is not as highly organized as that of wood and bone. Wood contains elongated pores (called tracheids or fibres) oriented along the tree or limb axis, radial channels called rays, and larger sap channels[12,47]. The cell walls are themselves fibrous and consist of oriented cellulose in a hemicellulose and lignin matrix. The alignment of the tracheids is favourable for resisting the prevailing forces in the tree[12]. Trabecular bone has a spongy structure. The struts or ribs in trabecular bone have a complex internal structure (Fig. 4) similar to that of the osteon in compact bone.

Synthetic cellular materials are used for applications such as cushioning, filtration, insulation and lightweight sandwich cores. They tend to have either a two-dimensional or a three-dimensional structure (honeycombs and foams respectively). In most synthetic cellular solids, there is only one size scale other than the atomic: that of the cells. Some synthetic open-cell polymer foams can have an unintentional further hierarchical structure, however, in that the ribs may contain 'microcells'[48]. Aerogels are gels in which the fluid phase is air rather than solvent; they have submicroscopic pores with a wide range of sizes organized in a hierarchical structure[49,50]. The microstructure depends on density, and the Young's modulus varies as $\rho^{3.8}$. Foams with negative Poisson's ratio[51] have an inverted (re-entrant) cell shape in which the cell ribs bulge inward rather than outward. Although they are not hierarchical in themselves, they may be used to make hierarchical composites in which the open space in the cells is filled with a compliant solid or with a foam of smaller cell size. If the filler is viscoelastic, the composite's viscoelastic response can be made large when the filler experiences a larger local strain than does the composite. By choosing the relaxation rates of the large-cell foam and the filler, it is possible to design materials in which Poisson's ratio increases or decreases[52] with time.

The first quantitative analysis of open hierarchical structures is that of Parkhouse[6], who even proposed a competition for the most structurally efficient hierarchical design. Parkhouse considered lattice, truss, honeycomb and tubular geometries, and showed that hierarchical structure can be used in the design of structural elements which, for a given compressive strength, are much lighter than elements with simple structure. The effect of damage on reliability in homogeneous solids, in structures with one scale size and in those with hierarchical structure were described[53] in connection with continuum models of structures. Ashby[54] has elucidated the freedom that a designer of load-bearing components has in choosing material properties, section shape, and with cellular and composite materials, microstructural degrees of freedom.

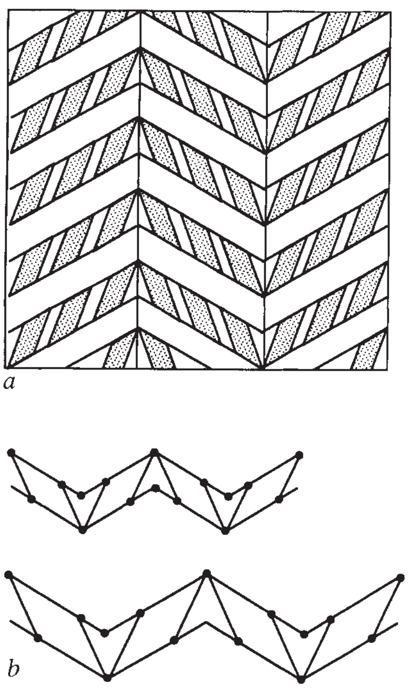

FIG. 3 *a*, Negative Poisson's ratio laminate of Milton[26]. *b*, Rod-and-hinge frame structure to illustrate the mechanics of the laminate in achieving a negative Poisson's ratio.

TABLE 1 Parameters in equations (1) and (3)

Microstructure	Stiffness Eq. (1) Exponent r	Stiffness Eqs. (1, 3) Multiplier k	Strength Eq. (3) Exponent q	Strength Eq. (3) Multiplier a
Foam, open cell, ribs bend[12]	2	1		
Elastic buckling		1	2	0.05
Plastic buckling		1	3/2	$0.3\varepsilon_{y,solid}$
Honeycomb (out of plane)[12]				
(Cell walls compress axially)	1	1		
Elastic buckling		1	3	5.2
Plastic buckling		1	5/3	$5.6\varepsilon_{y,solid}$
Brittle crushing		1	1	$12\varepsilon_{ult,solid}$
Ductile crushing		1	1	$\varepsilon_{y,solid}$
Frame (ribs deform axially)				
Elastic buckling				
oriented	1	1	2	0.05
cubic[9]	1	1/3	2	0.05
isotropic[9]	1	1/6	2	0.05

Prediction of strength and stiffness. In cellular materials, the stiffness depends on density and on the structure. In many such materials, which ordinarily have structure only on the scale of the cells, the relationships are simple. The Young's modulus (stiffness) E of a cellular material such as a foam or honeycomb (considered as a continuum) is given in terms of the Young's modulus E_0 of the solid from which the material is made, the density ρ_0 of the solid phase and the density ρ of the foam[12]. The atomic structure is ignored here because it is absorbed in the continuum description of the cell ribs.

$$E/E_0 = k[\rho/\rho_0]^r \quad (1)$$

The values of k and r depend on the type of structure (Table 1). In this section, stiffness and strength in hierarchical cellular solids is predicted for 'conventional' (not re-entrant) materials: honeycombs with hexagonal cells and for open-cell foams with convex (usually tetrakaidecahedral) cells. Simple continuum models now available for cellular solids[12] aid the analysis. Hierarchical cellular solids are considered in which the material making up the cell ribs is also cellular and has a smaller cell size. For a solid material viewed as a continuum, the hierarchical order $n = 0$; for a conventional foam or honeycomb, $n = 1$; and for a sponge with porous ribs, $n = 2$. Hierarchical solids may be envisaged with any hierarchical order, with an upper bound on n determined by the fact that the smallest cells must be of larger size than atomic dimensions.

To predict the properties of hierarchical cellular solids, one can iterate the stiffness equation (1), considering the solid density to be ρ_0 at zeroth order. Strictly, the classical continuum view used here is valid only if the size of the structure making up each cell wall or rib is much smaller than the rib itself.

$$E_n/E_0 = k^n[\rho/\rho_0]^r \quad (2)$$

On this basis, we see that for honeycombs deformed out of plane and for foams ($k = 1$), the hierarchical order n does not influence the stiffness, whereas for framework-type structures for which $k < 1$ the stiffness decreases with n. The relationships

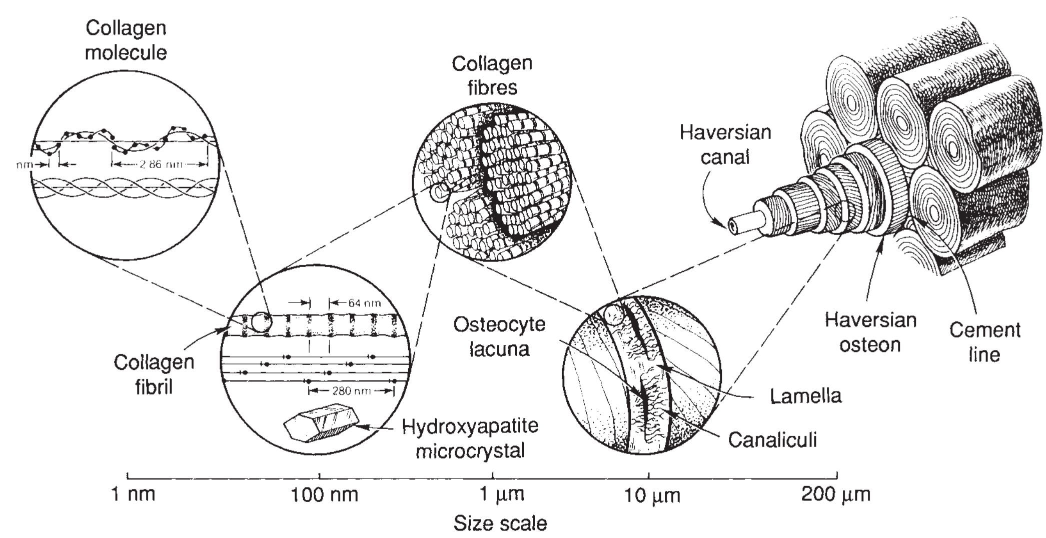

FIG. 4 Hierarchical structure in human compact bone; individual size scales adapted from refs 28–31. Fibrous, laminar, particulate and porous structure is present at different size scales.

for compressive strength[12] σ, or maximum stress before collapse, of conventional first-order materials ($n=1$ for cellular material, $n=0$ for the solid phase) can be rewritten in a general form for two arbitrary hierarchical orders differing by one, with $n \geq 1$. Here a and q are parameters which depend on the rib failure mode and material structure (see Table 1).

$$\sigma_n = akE_{n-1}[\rho_n/\rho_{n-1}]^q \qquad (3)$$

Possible failure mechanisms are elastic buckling of cell ribs in which the ribs reversibly collapse, plastic buckling in which the ribs irreversibly collapse, or crushing in which the ribs fracture. As shown in Table 1, the strength parameter a for plastic buckling depends on $\varepsilon_{y,\text{solid}}$, the strain at yield for the solid from which the material is made; for crushing, it depends on $\varepsilon_{\text{ult,solid}}$, the fracture strain of the solid. After some manipulation, and assuming the density ratio is the same for each level, we find that the strength to density ratio of the hierarchical material is, for $n \geq 1$

$$\frac{\sigma_n}{\rho_n} = \frac{E_0}{\rho_0} ak^n \left(\frac{\rho_n}{\rho_0}\right)^{r-1+(q-r)/n} \qquad (4)$$

The upper bound on the strength-to-density ratio is that of the solid at the zeroth level of the hierarchy. The crush limit for honeycomb is independent of hierarchical order: as $k=1$ and $q=r=1$, n has no effect.

The predicted strength-to-density ratio of honeycombs is shown in Fig. 5. The physical mechanism for the improved strength is the suppression of buckling in the hierarchical structure. Because failure can occur by elastic buckling, plastic buckling or crushing, the actual strength corresponds to the lowest stress of the possible failure modes. For low-density honeycombs, marked improvement in compressive strength can be realized in hierarchical structures. Most of the gain in strength occurs in the first few levels of the hierarchy; the situation for large n is one of diminishing returns. Moreover, the relative thickness of the cell walls increases with n, so for a relative density of 0.01, values of n above 4 are unrealistic. If the solid phase had a higher ultimate strain (for example $\varepsilon_{\text{ult}} = 0.054$ for glass fibres; 0.08 for silica) the transition from elastic buckling to plastic buckling in Fig. 5 would occur at higher stress. Consequently, there is a considerable advantage to using high-strength materials in making a hierarchical honeycomb, in contrast to conventional honeycomb in which only the stiffness of the solid phase is important.

As for foams, conventional open-cell foams deform by rib bending[12] for which $r=2$ and $k=1$ in equation (1). For such foams, there is no strength advantage associated with the hierarchical structure (Fig. 6). The situation is different for an oriented foam[12,55] such as trabecular bone of a particular structure (modelled as first-order) in which deformation proceeds by axial rib extension with $r=1$ and $k=1$ in equation (1). A cubic lattice of beams ($k=1/3$), more representative of building construction than of foams, deforms axially in this way. Such materials and structures are predicted to have a considerable strength advantage associated with the hierarchical structure (Fig. 6). For large n, the cubic and isotropic microstructures perform less well, because at each level, some ribs are oriented so that they do not support load.

These predictions of stiffness and strength are independent of scale and are relevant to large structures as well. If elastic buckling were the only failure mode considered (as in ref. 6), the saving in strength or weight of these structures would be overestimated.

A different type of analysis which generates hierarchical structure is finite-element analysis in which the topology of a structural element is allowed to vary[56]. In finite-element analysis, stress and deformation fields in an object are analysed by subdividing a model of the object into many small segments, for each of which it is simple to compute stress. In most applications

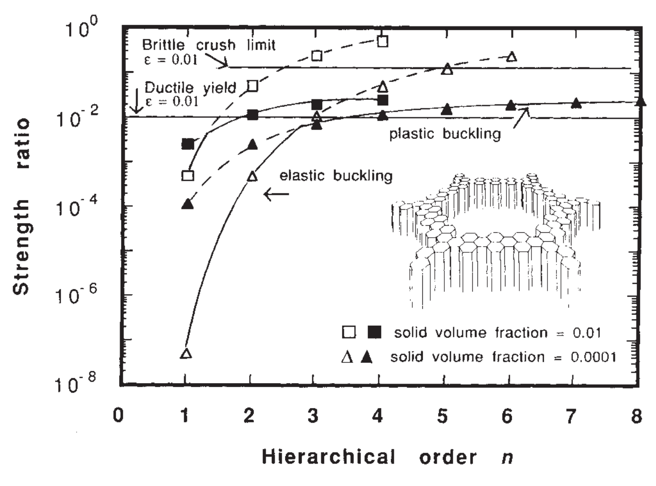

FIG. 5 Strength-to-density ratio of hierarchical honeycomb increases with hierarchical order n. Normalized strength ratio $(\sigma_n/\rho_n)/(E_0/\rho_0)$, for hierarchical honeycomb microstructure, as a function of n, for several solid volume fractions ρ_n/ρ_0: squares, 0.01; triangles, 0.0001. For the solid, the yield strain and fracture strain are assumed to be 0.01. Open symbols: elastic buckling. Solid symbols: plastic buckling. Inset: second-order honeycomb cell. Solid curves: buckling mode with lowest stress limits the strength.

of the method, the boundaries of the model are not changed. In ref. 56, holes were deliberately introduced in the model, and their boundaries were progressively modified depending on the calculated stress at each iteration. Optimization of the structural element to maximize stiffness for given weight leads to a cellular microstructure which becomes truss-like if the solid volume fraction is small. The optimal microstructure under some conditions is hierarchical.

Second-order hierarchical honeycombs can easily be made by the techniques currently used to make practical expanded honeycomb. In the 'hobe' (honeycomb before expanding) block method, strips of material are bonded together with bands of adhesive so that the bonded regions of one strip lie above the unbonded regions of adjacent strips. To produce a honeycomb, the width of glued and unglued sections is made equal. The stack of strips is pulled apart so that the web between the bonded strips forms the cell walls. I have used a modified hobe-block approach to make hierarchical honeycomb by stacking unexpanded small cell layers and using wide glue strips between them to make large cells with small cells in the walls. I also constructed hierarchical honeycomb by cementing together segments of first-order honeycomb to form a larger honeycomb. Experiments using a servohydraulic test machine to crush the specimens showed that second-order paper honeycomb is a factor of 3.2 to 3.8 times stronger in compression than first-order honeycomb of the same density (0.01 g cm^{-3}). The simple theory developed above, assuming plastic buckling, predicts a strength enhancement factor of 4.6. Given the idealizations involved, including identical density ratio at each order, this agreement seems reasonable. Honeycomb made of a stronger material would be enhanced even more in strength by hierarchical structure, as the transition between the governing failure modes depends on the strength of the material used.

Conclusions and prospects

Many materials exhibit hierarchical structure; the hierarchical aspects of structure are useful for descriptive purposes, for analysis and for synthesis. Hierarchical cellular material microstructures sometimes have far higher compressive strength than cellular solids of similar density with conventional structure. Two-dimensional hierarchical cellular solids (honeycombs) are easily made, and other hierarchical cellular solids could be manufactured by rapid prototyping systems[57] in which a computer-generated design is converted into complex shapes by photochemical, sintering, deposition, layering or sculpting techniques. Amongst the useful material properties that may be conferred by hierarchical structure, an intriguing possibility is that of simultaneously achieving values of strength and toughness, for which ordinarily there is a trade-off. Hierarchical structure can give extremal values of unusual properties such as negative Poisson's ratios. There is the further possibility of designing materials with extreme values of properties such as thermal expansion or piezoelectricity. □

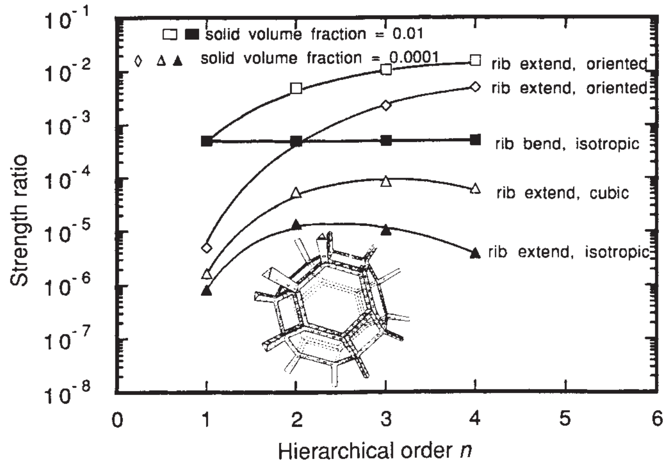

FIG. 6 Strength-to-density ratio of hierarchical foam can increase with hierarchical order n. Normalized strength ratio $(\sigma_n/\rho_n)/(E_0/\rho_0)$, for different types of hierarchical foam microstructure, as a function of n, for several volume fractions ρ_n/ρ_0. Solid is assumed to be sufficiently strong that failure occurs by elastic buckling. Inset: second-order foam cell, showing rib bend.

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