A triumph of lateral thought

ANDREW ALDERSON

Imagine stretching elastic and seeing it get fatter rather than thinner. It may sound bizarre, but this property is what makes auxetic materials potentially so useful

Materials can be divided into two basic categories: structural or functional. Development of structural materials is focused on improving their mechanical or physical properties, often with a saving in weight or cost. By contrast, functional materials are designed to detect and/or respond to events or stimuli that occur during their lifetime. These materials often display novel and counterintuitive behaviour. Examples include electrically (semi)conducting polymers, materials that contract when heated, and those that expand when subjected to hydrostatic pressure.

Another example is a remarkable class of materials known as auxetic materials.[1] When stretched lengthways, these materials get fatter rather than thinner (see Figure 1). As well as this unique characteristic, auxetic materials have enhanced mechanical and physical properties, which means that they can actually be classified as both structural and functional materials.

The key to auxetic behaviour is a value known as Poisson's ratio. This is defined as the ratio of the lateral contractile strain to the longitudinal tensile strain for a material undergoing uniaxial tension in the longitudinal direction. In other words, it determines how the thickness of the material changes when it is stretched lengthways. When an elastic band is stretched the material becomes thinner, giving it a positive Poisson's ratio. Indeed, most solids have a Poisson's ratio of around 0.2–0.4.

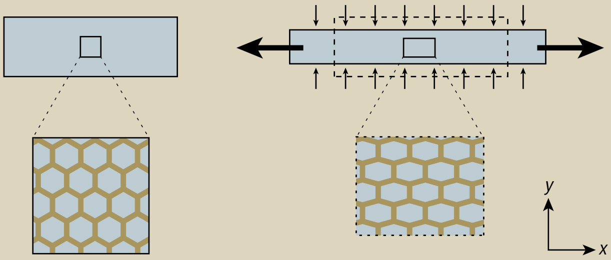
Poisson's ratio is determined by the internal structure of the material. For example, consider a two-dimensional honeycomb deforming by hinging of the ribs forming the network (see Figure 1). For the conventional hexagonal geometry (see Figure 1a), the cells get longer in the x-direction and close up along the y-axis when the material is stretched along the x-axis, giving a positive value for Poisson's ratio. Modifying the honeycomb cell geometry to adopt a 'bow-tie' structure (see Figure 1b) means that the network gets longer in both the x- and y-directions when it is stretched, giving it a negative Poisson's ratio and making the material auxetic.[2]

Auxetic materials are interesting both because of their novel behaviour and because of enhancements in other material properties that are related to Poisson's ratio. For example, hardness can be increased in an auxetic material (see Figure 2). When an object hits an auxetic material and compresses it in one direction, the auxetic material also contracts laterally — material 'flows' into the vicinity of the impact. This creates an area of denser material, which is resistant to indentation.

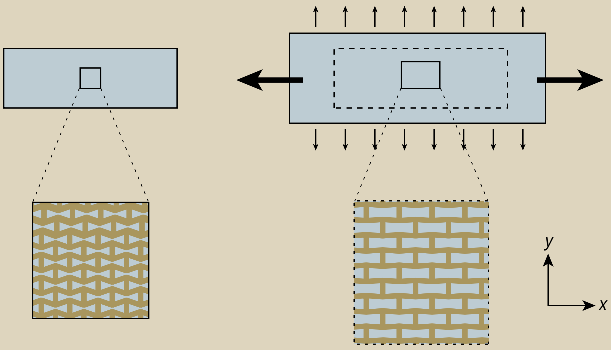
Importantly, elasticity — and hence auxetic behaviour — does not depend on scale. Deformation can take place at the macro-, micro- or even molecular level (see Figure 3). This means that we can not only consider auxetic materials, but also auxetic structures.

Auxetic behaviour **1**

(a) Non-auxetic material
As the material is stretched the component cells get longer in the x-direction but become compressed in the y-direction



(b) Auxetic material
As the material is stretched, the cells get larger in both the x- and y-directions

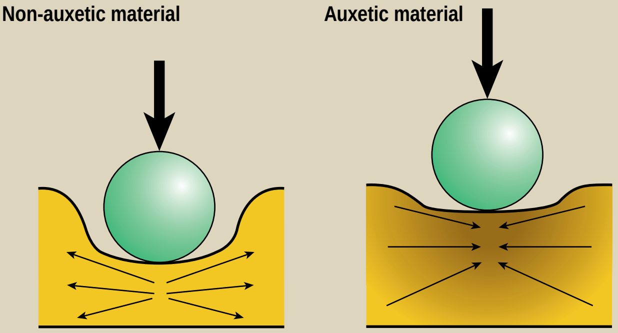


Thinking big

One of the largest examples of auxetic structures is the graphite core found in some nuclear reactors. These cores were developed in the late 1950s[3] and so pre-date the bulk of auxetic materials research by some 30 years or so. Indeed, these structures were not designed specifically to have auxetic properties. Instead, they were made to withstand the horizontal shear forces generated during earthquakes, while also allowing free move-

A harder material **2**

Auxetic materials are more resistant to indentations than ordinary materials. An auxetic material contracts laterally when hit by an object — material effectively flows to the site of the impact rather than away from it. This makes the auxetic material more dense at the site of the impact and therefore more resistant to indentation

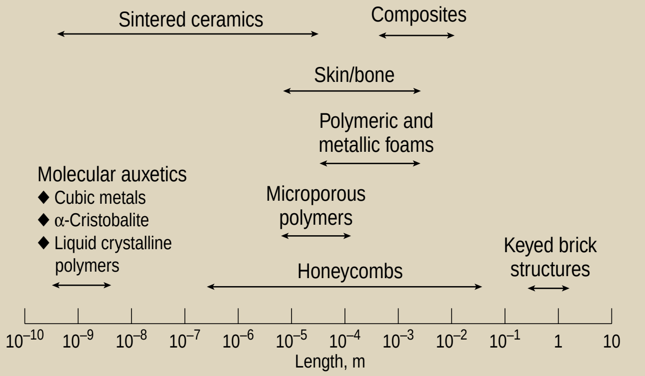

Non-auxetic material **Auxetic material**

ment of the structure in response to thermal movements between the graphite core and steel supporting structures, and expansion and shrinkage of the graphite during exposure to radiation. In other words, the structure had to have a high resistance to horizontal shear deformation and a low resistance to changes in volume.

A Magnox reactor core is made up of free-standing columns of graphite bricks, with central channels for the fuel and control rods. The bricks are connected by loose side and corner keys in keyways (see Figure 4). The structure expands in all radial directions when subject to a tensile load and, furthermore, retains its square lattice geometry during deformation. This makes the structure auxetic, with a Poisson's ratio of –1 in the horizontal plane. For isotropic materials and structures this value of Poisson's ratio corresponds to an infinitely high shear modulus with respect to the bulk modulus — exactly the properties required in the design stage. The auxetic properties were more a lucky result than a conscious part of the design, and it would be another two or three decades before the significance of this result would be fully appreciated and applied to other materials and structures.

A question of scale **3**

Auxetic materials and structures from the macroscopic down to the molecular level

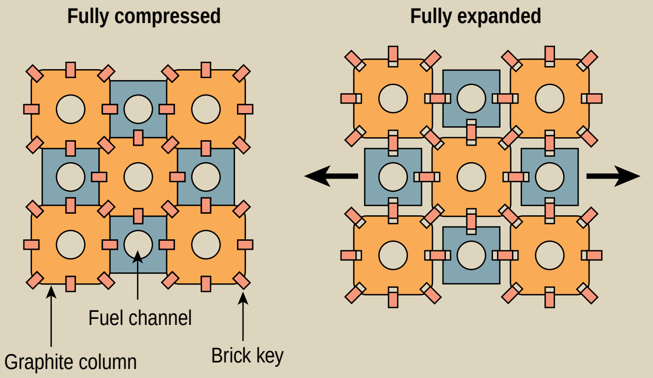


Calling in the cellular foams

Large-scale auxetic cellular structures were first realised in 1982 in the form of two-dimensional silicone rubber or aluminium honeycombs deforming by flexure of the ribs.[4] These structures are elastically anisotropic — that is, they have a different Poisson's ratio depending on the direction in which they are stretched. The current interest in auxetic materials really started with the development in 1987 of isotropic auxetic foams by Roderic Lakes of the University of Iowa (now at the University of Wisconsin-Madison).[5] Polymeric and metallic foams were made with Poisson's ratios as low as –0.7 and –0.8, respectively.[6,7] Whereas conventional foams are made up of convex polyhedral cells, these new auxetic foams feature much more convoluted cell structures (see Figure 5).[8]

Large scale auxetics **4**

The keyed brick structure of a Magnox reactor core displays auxetic behaviour on a macroscopic level. The structure expands in all radial directions when subjected to a tensile load.



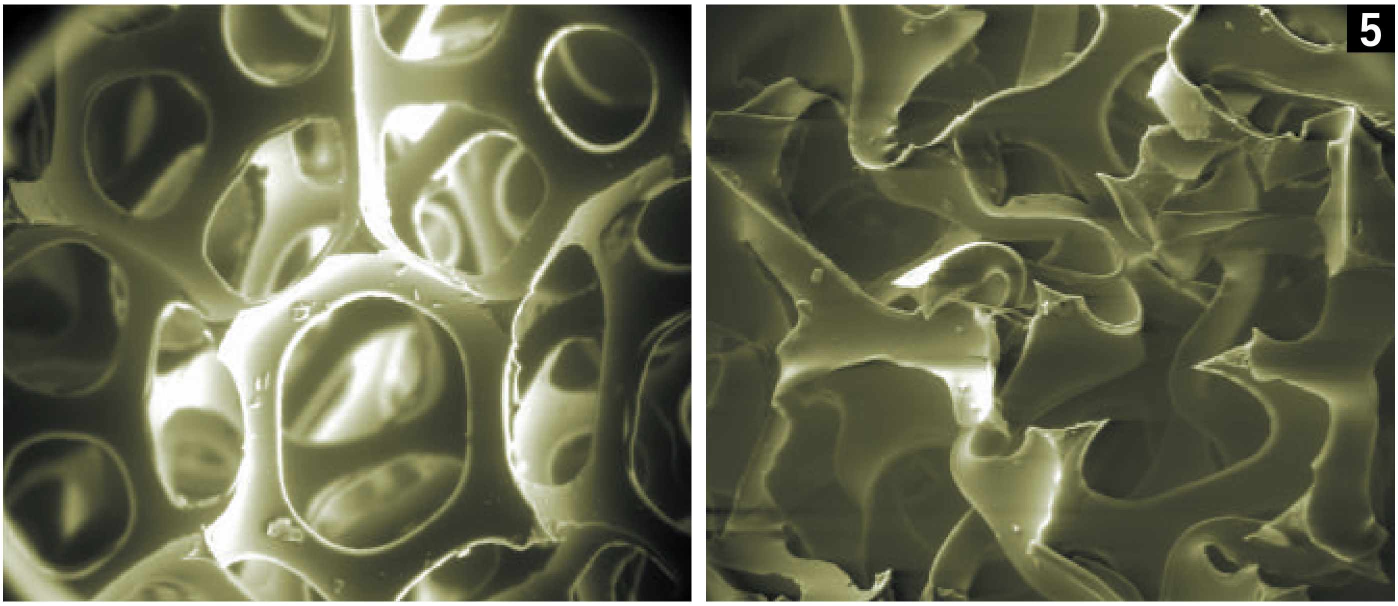
Foams have a variety of uses — in packaging, sound insulation, air filtration, shock absorption and as sponge materials, for example. A range of properties have been studied for auxetic foams. Lakes found that auxetic foams are more resilient than non-auxetic materials.[5] In addition, when they are subjected to a bending force auxetic foams undergo double curvature into a dome-like shape, rather than forming the saddle shape adopted by non-auxetic materials.[9] Both of these factors could be important in cushion materials. Resilience is related to comfort, and the double curvature may be useful in ensuring mattresses, for example, provide optimal support for the 'doubly curved' human body.[10]

Dynamic effects have also been investigated[11] and studies partially funded by the US Office of Naval Research[12] have shown that auxetic foams are better than their non-auxetic counterparts at absorbing sound and vibration. Lakes has also used copper foam as an auxetic press-fit fastener. The fastener is easy to insert because it contracts radially in response to the applied pressure, and it resists extraction by pulling because of radial expansion.[13] Other studies, with support from NASA and Boeing, have demonstrated enhancements in shear resistance,[7,14] indentation resistance[14,16] and fracture toughness for the foams.[17]

Lakes has now developed processing techniques to make larger auxetic foam 'slabs'.[18] Ken Evans of Exeter University has also developed a multi-stage processing route for large auxetic foam blocks,[19] with improved process control enabling more homogeneous and stable foams to be made, as well as both isotropic and anisotropic ones. These foam blocks should find applications in mattresses and wrestling mats, for example.

When foams are subjected to stress, their permeability varies as the cells are distorted. A recent collaboration between British Nuclear Fuels plc (BNFL) and Evans' group found that this variation in permeability is enhanced in auxetic polymeric foams and honeycombs with cell dimensions of around 1mm.[20] This makes them potentially useful for filtration applications — they afford greater control of the pressure existing across the filter, as well as enabling particulate defouling when a load is applied. However, pores smaller than 1mm are needed for these benefits to be realised in many practical filtration applications.

Methods for scaling down honeycomb-like cellular structures include LIGA technology, laser stereolithography,[21] molecular self-assembly,[21] silicon surface micromachining techniques[22] and nanomaterials fabrication processes.[23] Auxetic two-dimensional cellular structures with cell dimensions of about 50μm have been made by Ulrik Larsen and co-workers at the Technical University of Denmark,[22] and three-dimensional microstructures consisting of two-dimensional conventional and auxetic honeycomb patterns on cylindrical substrates have recently been designed and made by George Whitesides and co-workers at Harvard University.[24] These have potential in micro- and nanotechnology applications (for example, in compliant micrograppers or micropositioners used in fields such as microsurgery and nanofabrication).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Secrets of the cells: auxetic (right) and non-auxetic open cell foam pore structures

Polymeric improvements

In general, foams are simply not stiff enough to use as structural materials. Hence, further development in auxetic materials sought to design and make stiffer materials for wider engineering applications. In 1989, in research performed at Liverpool University, Brian Caddock and Evans discovered that an expanded form of microporous polytetrafluoroethylene (PTFE) was auxetic.[25]

The auxetic behaviour of the expanded PTFE is a result of the particular microstructure formed rather than any intrinsic property of PTFE itself.[26] The microstructure consists of an array of particles ('nodules') interconnected by fibrils, analogous to the honeycomb structure, where the vertical and diagonal honeycomb ribs correspond to the PTFE nodules and fibrils, respectively.

In further research at Liverpool University, Evans and Kim Alderson, funded by the British Technology Group, developed a processing route for auxetic ultra-high molecular weight polyethylene (UHMWPE)[27] with a similar nodule–fibril microstructure. The stiffness of this polymer is an order of magnitude greater than that of auxetic foams, and is comparable to conventional thermoplastic polymers.[28] Auxetic UHMWPE has improved indentation resistance[29] and attenuation of ultrasonic signals.[30]

Subsequent refinements in processing[31] (partially funded by ICI Chemicals and Polymers) and increased understanding of the deformation processes responsible for the strain-dependent behaviour[32,33] mean that the production of auxetic polymers with specifically tailored properties is now a real possibility. Auxetic polypropylene has now been made,[34] and the requisite microstructure has also been produced in nylon[32] and most recently, in work by Kim Alderson and co-workers at Bolton Institute, in polypropylene fibres.[35]

Following on from examples of naturally occurring auxetic biomaterials, which include cow teat skin[36] and cat skin,[37] man-made auxetic polymers may find useful applications in medicine. For example, Evans and Caddock have studied the properties of expanded PTFE and fibrillar polyurethane (PU) arterial prostheses.[38] They found the two to be markedly different — PTFE was auxetic while PU was not. This has implications for the performance of the two prostheses. An auxetic prosthesis may be a better match to the properties of natural biomaterials. In addition, in a non-auxetic vessel, a pulse of blood may cause the wall of the vessel to rupture as a result of thinning, whereas the auxetic vessel's wall would actually thicken and thus resist rupture.

A strong combination

Composite materials are made up of two or more different components to give properties that are superior to those of the individual components. Composites typically have high strength- and stiffness-to-weight ratios, making them useful in, for example, aerospace and automobile applications. Composites offer a route to auxetic materials of higher stiffness than auxetic microporous polymers.

Evans' group and Ian Ward's group at Leeds University have configured carbon-fibre-reinforced epoxy composite laminate panels so that they are auxetic.[39] These are a further two orders of magnitude stiffer than auxetic UHMWPE. They also have enhanced fracture toughness and indentation resistance.[39] This is significant because composite laminates are usually damaged relatively easily by low load indentation.

With funding from Ciba-Geigy, auxetic composite sandwich panels have been made from an auxetic honeycomb core material, usually aluminium or resin, bonded to outer surface layers of a fibre-reinforced composite laminate material.[40] These panels

Piezoelectric auxetics

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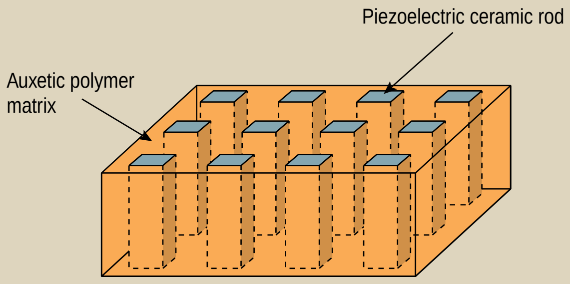
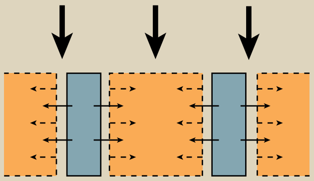
Basic structure of a piezoelectric composite using an auxetic matrix



When compressed, the radial expansion of the non-auxetic ceramic rods is helped by the radial contraction of the auxetic polymer matrix



can be formed into doubly curved or domed shapes as a result of the curvature properties of auxetic materials. This eliminates the need for the expensive and damaging machining techniques used to shape non-auxetic panels. Applications for these sandwich panels include curved body parts for cars and aircraft. Further work on modifying and enhancing auxetic composite designs should result in even stiffer composites.[41–43]

Piezoelectric composites — which convert a mechanical stress into an electrical signal and vice versa — consist of piezoelectric ceramic rods within a passive polymer matrix (see Figure 6). They are used in medical ultrasonic imagers and naval sonar receivers. The characteristics of the device depend on the matrix properties. In recent designs by Wallace Smith of the US Office of Naval Research,[44] the sensitivity of a sonar receiver was increased by an order of magnitude by replacing a non-auxetic matrix with an isotropic auxetic matrix. Further improvements are predicted for a highly anisotropic auxetic matrix.[45] Steps are already being taken towards the practical realisation and testing of these devices. Laser stereolithography has been used to develop a prototype of a three-dimensional unit-cell designed to give optimal performance as a result of auxetic behaviour.[46]

Ceramics: fighting fractures

But it is not only the polymer matrix in composite materials that could benefit from the use of auxetic material. The manufacture of auxetic ceramics is also under investigation. Typically, ceramics are made by compacting powders or particles into solid ‘green compacts’, which are subsequently heated to bond the particles together (‘sintering’). Additives such as binders to promote bonding of the particles may be included in the process. The porosity and final size of the particles (or grains) and, therefore, the mechanical properties of the product can be controlled, although fractures and microcracks are inevitably introduced during this process.

By introducing the auxetic effect, fracture toughness could be improved, leading to high-performance ceramic components. However, ceramics are usually less porous than the cellular and microporous auxetic materials, so an alternative mechanism for auxetic behaviour is likely to be needed to realise auxetic ceramics.

In fact, auxetic sintered ceramics have been reported in certain bismuth cuprate superconducting polycrystalline compounds.[47] However, the mechanism responsible for the auxetic effect is not clearly understood, although auxetic behaviour has been predicted for bonded granular materials.[48]

In an alternative approach, Hassel Ledbetter and co-workers at the National Institute of Standards and Technology in Colorado have investigated the effect of porosity and microcracks in elastic solids.[49] They suggest that a solid with a porosity of less than 40% cannot be auxetic unless the solid is intrinsically auxetic at the molecular level. However, in another approach, auxetic behaviour is predicted in solids with cracks, even for an intrinsically non-auxetic material.[50] It is known that some porous rocks are auxetic, which may support this latter view.[51]

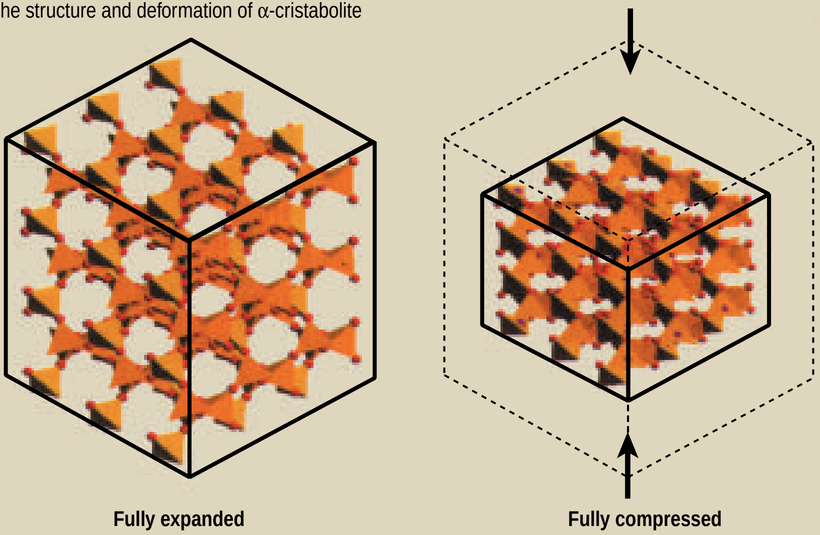
Molecular auxetics

The drive to design and synthesise new auxetic molecular materials arises partly from the desire to make materials with extreme properties, and partly from the novel properties and applications of stable auxetic coordination solids containing molecular-sized cavities and pathways. A rich variety of host–guest chemistry can be envisaged with potential in sensor, molecular sieve and separation technologies, for example.

Ledbetter has found evidence of auxetic behaviour in $YBa_2Cu_3O_7$,[52] and so auxetic behaviour at the molecular level is thought to exist in some oxide superconducting compounds. Some naturally occurring single crystal materials such as arsenic[53] and cadmium[54] also exhibit auxetic behaviour. Studies in 1979 and, more recently, by Ray Baughman and co-workers at AlliedSignal in 1998 have revealed that 69% of the cubic elemental metals and some rare gas solids are auxetic when stretched along a specific direction.[55,56] Baughman has further suggested that auxetic metal electrodes would give a two-fold increase in piezoelectric device sensitivity. Recently, the α-cristobalite polymorph of crystalline silica was found to be auxetic.[57] The mechanism behind this effect is probably rotation of the SiO_4 tetrahedral units making up the α-cristobalite molecular structure (see Figure 7).[57,58]

Molecular auxetics **7**

The structure and deformation of α-cristobalite



Auxetic behaviour has been predicted for molecular networks based on the familiar honeycomb geometry.[1,59] Baughman has proposed alternative molecular networks, called ‘twisted-chain auxetics’. Here, auxetic behaviour is a result of a specific shear deformation in helical polyacetylene chains formed from adjacent chains in a coupled polydiacetylene chain network.[60] These twisted-chain auxetics are likely to have a range of useful properties including contraction when heated, expansion under pressure, a semiconductor-to-metal transformation when exposed to dopants, and shape-memory behaviour. Furthermore, these twisted chain auxetics may have interesting electrical and optical properties, offering potential for displays and electromechanical actuators.

Molecular modelling calculations performed by Yuejin Guo and William Goddard III at the California Institute of Technology have predicted that the α and β phases of carbon nitride are auxetic.[61] This could be important as carbon nitride is a leading candidate in the search for materials harder than diamond.

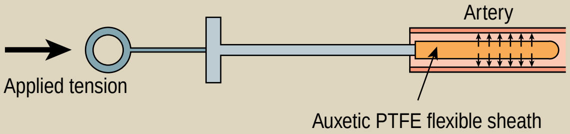
Nanoscale macrocyclic hydrocarbons similar to the conventional molecular honeycomb network sub-units proposed by Evans have been synthesised by Jeffrey Moore of the University of Illinois,[62] although no progress on the auxetic sub-units has so far been reported. Moore has also successfully synthesised hinged coordination networks similar to Baughman's twisted-chain auxetics, although the mechanical properties do not appear to have been measured.[63] Recently, however, in research funded by the US Air Force Office of Scientific Research, auxetic behaviour has been realised in a main-chain liquid crystalline polymer synthesised by Andy Griffin's group at the University of Southern Mississippi.[64] Each polymer chain consists of a series of rods interconnected by flexible 'spacers'. The rods are connected terminally or laterally, in an alternating fashion, by the spacer groups. In one phase — the nematic phase — all the rods are oriented along the chain direction. Auxetic behaviour occurs when the chain is stretched because the laterally attached rods rotate perpendicular to the chain, causing an increase in the lateral inter-chain separation. Griffin describes these liquid crystal polymers as having 'interesting and potentially useful physical properties'.[65]

Auxetics in action

8

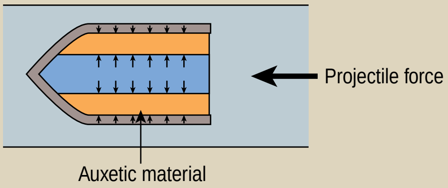
Artery dilator

Applying tension to the auxetic sheath causes it to expand laterally, which opens up the artery



'Bullet'

The force pushing the projectile down the barrel radially compresses the auxetic material, rather than causing sideways expansion



Expanding the horizons

Auxetic materials research has now reached the stage where an increasing number of materials and processing routes are being developed and specific applications are being addressed. With further progress in the fabrication and synthesis of a wider range of these exciting materials there is enormous potential for application in industrial and commercial sectors.

In addition to the examples already mentioned, there is a small but growing patent portfolio relating to auxetic materials. For example, a dilator for opening the cavity of an artery or similar vessel has been described for use in heart surgery (angioplasty) and related procedures (see Figure 8).[66] The coronary artery is opened up by the lateral expansion of a flexible auxetic PTFE hollow rod or sheath under tension. Another biomedical application relates to auxetic surgical implants.[67]

Toyota has recently patented a manufacturing route for auxetic composites[68] and a drive unit for feed gear rotation formed from auxetic material.[69] Auxetic fibre-reinforced composite skis, with a lower resistance to motion, have been described in a patent by Yamaha.[70] Mitsubishi has patented a 'narrow passage moving body with highly efficient movement' — in other words a bullet — in which one component is made of an auxetic material so that the overall object has a Poisson's ratio of zero (see Figure 8).[71] In this case the movement of the projectile down a barrel, for example, is helped because the sideways expansion arising from the thrusting force is reduced. Auxetic materials have also been identified as candidate materials for use in electromagnetic launcher technology to propel such projectiles.[72] And the intended recipient of the projectile might benefit from a bullet-proof vest and other personal protective equipment formed from auxetic material because of their impact property enhancements. Here, the Defence Clothing and Textile Agency in Colchester has been looking into the use of auxetic textiles for military purposes.[10] It may be possible to produce auxetic body armour that is both lighter and thinner than conventional body armour.

Of course large-scale auxetic structures are already in use in the form of the nuclear reactor cores. Commercially available auxetic materials in use include pyrolytic graphite for thermal protection in aerospace applications,[73] and large single crystals of Ni_3Al in the vanes of aircraft gas turbine engines.[56] However, it is reasonable to assume that these materials and structures were not (knowingly) deployed because of their auxetic properties. In order to develop a wider range of auxetic materials and structures it will be necessary to maintain a multidisciplinary research effort, as might be expected for a class of materials/structures spanning construction engineering to molecular engineering. Biomedical and nanotechnology applications are particularly exciting examples of potential end uses for auxetic materials.

Similarly, the combination of the auxetic effect with other novel effects, such as in Baughman's auxetic molecular networks, should lead to a rich vein of material functionality. Baughman has also suggested applications for materials that expand when put under pressure, and notes that there are examples of crystals demonstrating both this property and auxetic behaviour.[74]

There are significant challenges pertaining to supramolecular chemistry as we strive to develop auxetic materials at the molecular level. However, given the pace of progress in these and other areas, it is clear that auxetic materials have a key contribution to make in the development of new and improved structural and functional materials. They are indeed examples of new materials from lateral thinking — literally.

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