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Workings of auxetic nano-materials

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ABSTRACT

Purpose: The human mind is consistently interested in new materials having unique properties. Recently, a relatively new field is being investigated which exhibits a negative Poisson's ratio (NPR), and consequently are termed auxetic materials.

Design/methodology/approach: One of the main reason for interest in auxetic materials is due to the possibility of enhanced mechanical properties such as shear modulus, plane strain fracture toughness and indentation resistance compared to non auxetic material.

Findings: Auxetic materials were described concerning their classification, characteristic, properties and potential applications.

Research limitations/implications: The paper is an overview the modelling structure and deformation mechanisms of auxetic nano-materials.

Originality/value: The paper shows the possibilities of auxetic materials application resulting from their mechanical properties.

Keywords: Auxetic; Nano-materials; Negative Poisson'a ratio

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1. Introduction

Auxetic materials are typically divided into two classes in terms of their scale, which are the common auxetic materials (macro-scale) and nano-scale auxetic materials. This paper, mainly concentrates on study of materials having auxetic behaviour at the molecular level and investigating their deformation mechanisms.

When a material is stretched in one direction, it tends to contract (or, rarely, expand) in the other two directions. Conversely, when a sample of material is compressed in one direction, it tends to expand (or rarely, contract) in the other two directions. A measure of this dimensional change can be defined by Poisson's ratio (ν),

$$\nu_{yx} = -\frac{\text{strain in the lateral direction}}{\text{strain in the loading direction}} = -\frac{\varepsilon_x}{\varepsilon_y} \tag{1}$$

where ε_x and ε_y are the strains in the x and y directions, respectively.

In fact, for most materials this value is positive (Poisson's ratios ranging between 0 and 0.5) and reflects a need to conserve

volume. Negative Poisson's ratio (NPR) material expands (or contracts) laterally when stretched (or compressed), in contrast to ordinary materials. These new types of materials were named "*auxetic*" by Evans [1]. "*Auxetic*" comes from the Greek word *auxetos*, meaning "that which may be increased".
Consider, as an example, auxetic ultra high molecular weight polyethylene (UHMWPE) proposed and fabricated by Alderson & Evans, 1992 to give a general idea as to what is the difference between auxetic and conventional material and to visualize the deformation mechanism (See Fig. 1) [2].

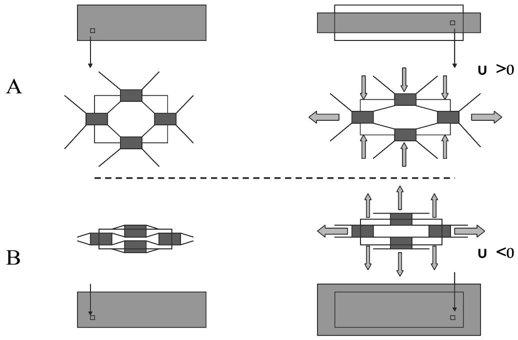


Fig. 1. Schematic diagram of structural changes observed in microporous UHMWPE undergoing tensile loading in the longitudinal direction: Non-auxetic (A) and auxetic (B) deformation due to fibril hinging in a nodule-fibril microstructure [2]

Fig. 1(a) on the top shows schematically the geometry and deformation of a common material undergoing lateral contraction for loading with a tensile longitudinal stress. Fig. 1(b) on the bottom shows auxetic behaviour in which the undeformed material (left) responds to the tensile longitudinal stress with lateral expansion (right).

2. Auxetic materials properties

Recently, the investigation of auxetic materials has attracted significant attention. One of the reasons for interest in auxetic materials comes from the fact that negative Poisson's ratio can lead to enhancement in other mechanical properties, including:

- Shear modulus [3,4],
- Indentation resistance [5,6],
- Dynamic properties [7,8],
- Fracture toughness [9].

Shear modulus

Auxetic materials have higher resistance to shear strain, which can be qualitatively explained by the relationships between shear (or rigidity) modulus G, Young's modulus E, bulk modulus K (the inverse of the compressibility) and Poisson's ratio ν. For isotropic materials the relationships between these constants are:

$$\nu = \frac{1}{2}\left(\frac{3K-2G}{3K+G}\right) \qquad (2)$$

$$G = \frac{E}{2(1+\nu)} \qquad (3)$$

$$K = \frac{E}{3(1-2\nu)} \qquad (4)$$

$$E = \frac{9KG}{3K+G} \qquad (5)$$

The Poisson's ratio of a stable isotropic material cannot be less than -1.0 or greater than 0.5 due to the requirement that the Young's modulus, shear modulus and bulk modulus have positive values [10]. However, the Poisson's ratio can exceed that limits in certain direction for anisotropic materials [11].

For isotropic materials, when Young's modulus (*E*) remains unchanged, the values of the shear modulus (*G*) and the bulk modulus (*K*) can be altered through the changes in Poisson's ratio (ν) (Eqs. 3 and 4). *E* is at least twice the value of *G* for positive Poisson's ratio materials. Conversely, *E* reduces to below 2*G* when ν is negative. For example, $E=G$ when $\nu=-0.5$ and $E\to 0$ when $\nu\to -1$. In other words, when decreasing ν to -1, the shear modulus tends to very high values and the Young's modulus decreases to low values, which makes a solid difficult to shear but easy to deform. *K* also decreases as $\nu\to -1$, which means the material becomes hard to shear but highly compressible. When ν approaches +0.5, the shear modulus (*G*) is greatly exceeded by the bulk modulus (*K*), which makes the material incompressible but easy to shear.

Indentation response

For isotropic materials, the indentation resistance (or hardness) (*H*) is inversely proportional to $(1-\nu^2)$ for a given pressure, defined as:

$$H \propto \left[\frac{E}{(1-\nu^2)}\right]^{\gamma} \qquad (6)$$

where the value of γ relates to the theoretical analysis used. $\gamma = 1$ stands for uniform pressure distribution [12] and $\gamma = 2/3$ is for Hertzian indentation [13].

Eq. 6 shows the indentation resistance tends to infinity with increase in the magnitude of Poisson's ratio (ν) for a given value of Young's modulus (*E*). As already noted, the range of Poisson's ratios for isotropic materials is $-1< \nu <0.5$, and so auxetic isotropic material show enhancements in indentation resistance when $-1< \nu < -0.5$. As ν approaches -1, the indentation resistance towards infinity. Enhanced indentation resistance of auxetic materials has been demonstrated through investigation of synthetic auxetic materials (i.e. polymeric and metallic foams [5,6]; and microporous polymers [14]). A schematic of indentation response is illustrated in Fig. 2 for both non-auxetic and auxetic materials subjected to compressive impact loading [15].

On the non-auxetic material, the force compresses the material in the direction of impact and the material spreads in directions perpendicular to and away from the direction of the impact (arrows define the lateral ‘flow’ of material). For the auxetic material, on the other hand, the vertical impact compression means that the material also contracts laterally-material ‘flows’ into the vicinity of the impact, leading to increased densification, therefore, enhanced indentation resistance [15].

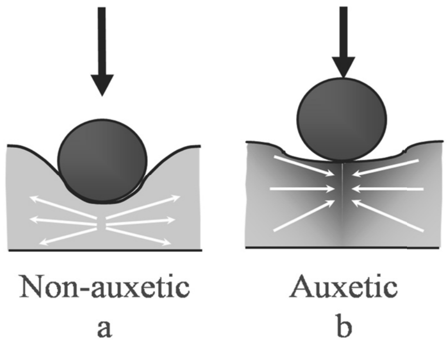


Fig. 2. An object hitting a non-auxetic (a) and an auxetic (b) material in the vertical direction. The arrows indicate the lateral ‘flow’ of materials in each case

3. Auxetic materials potential applications

3.1. Classifications and types

Within the last thirty years, a variety of auxetic materials and structures have been discovered, designed, manufactured or synthesised, ranging from the molecular level up to macroscopic level. Fig. 3 shows that there are examples of each of the major classes of materials (polymers, composites, metals and ceramics) with auxetic properties, and that natural and man-made auxetic materials are known over several orders of magnitude of stiffness, or Young’s modulus [16].

Auxetic materials have been discovered, fabricated and synthesized, such as polyurethane and polyethylene foams [17,18], microporous polytetra-fluoroethylene (PTFE) [19] microporous ultra high molecular weight polyethylene (UHMWPE) and polypropylene (PP), highly anisotropic composites [20], laminates [21,22], several types of rocks with microcracks [23,24], and naturally occurring molecular auxetics, e.g. α –cristobalite [25,26], and zeolites [27].

Applications

As stated in the above section, auxetic materials have demonstrated enhancements in other physical properties. Therefore, a range of potential applications of auxetic materials can be envisaged.

For example, auxetic foam with a re-entrant microstructure through thermomechanical processing was established to provide dispersion of acoustic waves and cut-off frequencies, which might have potential applications in the absorption of sound [18,29].

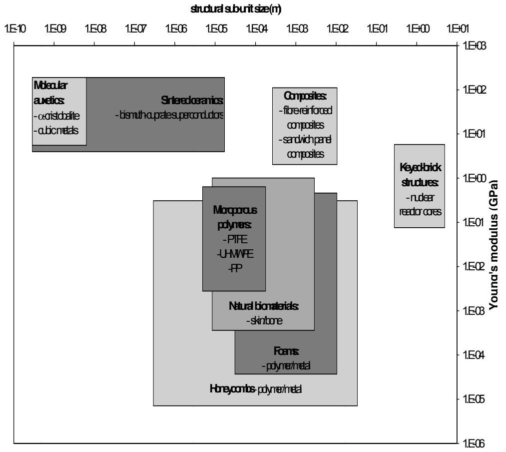


Fig. 3. Structured feature size and Young’s modulus of Auxetic materials from the macroscopic down to the molecular level [28]

Auxetic cellular solids (i.e. auxetic foams) could be used as fasteners and rivets due to the negative Poisson’s ratio. Insertion of the auxetic fastener is facilitated by the lateral contraction of the auxetic material under compression, while removal of the fastener is resisted since the fastener expands and locks itself more tightly in the hole under tension [30]. The scaling up of the processing of auxetic foam [31] opens up the development of seat cushions as another important potential application for auxetic foams. Wang and Lakes 2002 reported auxetic cushions could be beneficial in the prevention of pressure sores or ulcers in the sick and in reduction of pressure-induced discomfort in seated people [32]. Auxetic foam could also be used in ear cushions (US Patent US6412593 2002) for earphone shells due to their excellent features in forming synclastic doubly curved surfaces (reducing air leaks) and sound absorption property [33,34].

Alderson et al. 2000 reported auxetic materials have potential application in filters from the macro-scale (e.g. auxetic cellular structures with re-entrant honeycomb and auxetic foam) to the nano-scale (e.g. zeolites), which could improve filter cleaning and particulate size selectivity capabilities due to the unique pore variation properties of auxetic materials wide mechanical deformation [16].

Huang & Blackburn 2001 proposed the thermal shock resistance of ceramics with cellular or honeycomb structures could be enhanced by having negative Poisson’s ratios. In this case, they have applications as substrates in catalytic converters for the emissions from internal combustion engines. The relatively low bulk modulus associated with auxetic materials makes them more sensitive to hydrostatic pressure, and so, Avellanads, 1998 pointed out auxetic composites could be used in the design of hydrophones and other sensors. For example, piezocomposites consisting of piezoelectric ceramic rods within a passive polymer matrix and used in medical ultrasonic imagers and hydrophone receivers of naval sonar were shown to lead to order of magnitude enhancement in sensitivity when the polymer matrix assumed auxetic properties [35,36,37].

For fibre reinforced composites, fibre pull-out is a major failure mechanism. Auxetic materials are also predicted to enhance the failure resistance properties of composites. Alderson et al. 2002 reported the successful development of a melt-extrusion process to produce the first auxetic polymeric (polypropylene) fibre. It was postulated that composites with auxetic fibres could display reduced fibre pull-out, potentially helping to resist crack growth, through maintaining the interface by careful matching of the Poisson's ratios of the matrix and fibre expansion during loading [30,38,39]. Subsequently, comparative single-fibre pull-out test were performed using positive and negative Poisson's ratio polypropylene fibre and enhancement in maximum load (factor of 3 increase) and energy adsorption (factor of 3 increase) demonstrate for the auxetic fibre systems [40].

There are many potential applications for auxetic fibres in composite materials, such as vehicle body or car bumper; and in combination with other materials for personal protective clothing or equipment, e.g. in a crash helmet, projectile-resistant or bullet proof vest, shin pad, knee pad or glove. Other applications postulated for auxetic fibres include fishnet, replacement biomaterial, a bandage or wound pressure pad, and as a seal or gasket.

3.2. Limitations

As mentioned above, auxetic materials potentially have a wide range of applications and are attracting interest due to their unique behaviour. However, most manmade auxetic materials derive their behaviour from structure at micro or macro levels and so are less stiff than the solids from which they are made. Most auxetic materials have intrinsic porosity in order to achieve the negative Poisson's ratio (i.e. auxetic materials need space to allow "hinges" to flex, or "nodules" to spread out and so on). Therefore, many auxetic materials have limited potential in load-bearing applications.

A further reason for developing an understanding of the mechanisms responsible for auxetic behaviour at the nano-scale is for the development of auxetic nano-fibres to realise the already established benefits for auxetic macro-fibres in high performance composites containing auxetic nano-fibre.

3.3. Auxetic materials

Auxetic materials at the molecular level can be divided into two categories: natural and synthetic nanoauxetics. These will be broadly discussed in the following.

Molecular level auxetics I: Natural nanoauxetics

A negative Poisson's ratio was first reported in single crystal iron pyrites [41] as a result of crystals having a tightly constrained regular geometric arrangement and a twinned crystal arrangement. Since then negative Poisson's ratios have been reported in other single crystal phases. For example, carbon nitride has been suggested to have a negative Poisson's ratios [42]. It is believed that carbon nitride would be harder than diamond.

Baughman & Galvao 1993 have also found that a negative Poisson's ratio is very common in cubic crystals when they are stretched along the [110] and $[1\bar{1}0]$ direction. The deformation mechanism is illustrated in Figure 4. Cubic elemental metals were stretched along the [110] direction (axis between atom 2 and 4), causes atoms 2 and 4 to separate along [110], and atoms 1 and 3 to move closer together along the [001] direction (axis between atom 1 and 3). This then corresponds to extension and contraction along the [110] and [001] axes and so Poisson's ratio is positive. However, the moving together of atoms 1 and 3 along the [001] axis cases atoms 5 and 6 to move apart along the $[1\bar{1}0]$ axis due to the ribs connecting these atoms aligning towards the $[1\bar{1}0]$ direction. Hence, there is an expansion of the $[1\bar{1}0]$ dimension of the structure, accompanying the expansion in the [110] direction, leading to a negative value for Poisson's ratio [43].

For transversely isotropic zinc (with hexagonal monocrystalline structure), the auxetic behaviour has been found in its basal plane [44]. An orthorhombic alloy with auxetic behaviour is predicted by Rovati 2003. Meanwhile, Rovati 2004 also pointed out that 25% of monoclinic crystals displaying auxetic behaviour based on theoretical analysis [45,46].

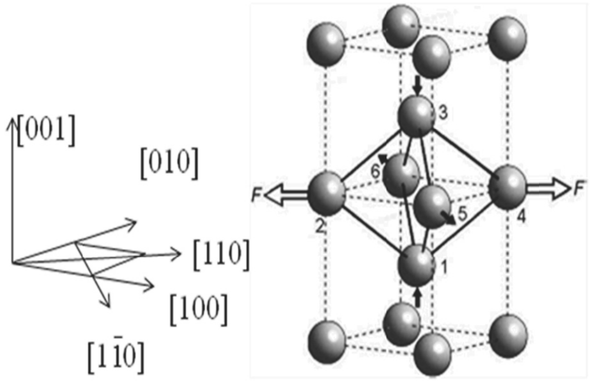


Fig. 4. The nanostructure of cubic metals with negative Poisson's ratios. (001) refers to the direction along a cubic principal axis [43]. [110] and $[1\bar{1}0]$ are directions at a 90° angle from a cubic principal axis

The main breakthrough in inorganic auxetics occurred when Yeganeh-Haeri et al 1992, measured abnormally high negative single crystal Poisson's ratios (up to -0.5 for some directions) in α–cristobalite. Fig. 5 shows that α–cristobalite consists of a framework of corner-shearing SiO_4 tetrahedron.

The single-crystal α–cristobalite elastic constants are highly anisotropic and by employing suitable averaging techniques [41], it has been calculated that an isotropic polycrystalline aggregate would also be auxetic. Negative Poisson's ratios also have also been observed in α-quartz, which also possesses a corner-shearing framework of SiO4 tetrahedral (See Fig. 6) as reported by Keskar and Chelikowsky 1992 [47].

Auxetic behaviour in α–cristobalite is thought to be due to the concurrent dilation and cooperative rotation (about tetrahedral axes in the x-y plane, passing through the midpoints of opposing edges) of the SiO_4 molecular tetrahedral making up the framework structure. Based on this ideal, an analytical model has been also developed for α–cristobalite and extended to α–quartz.

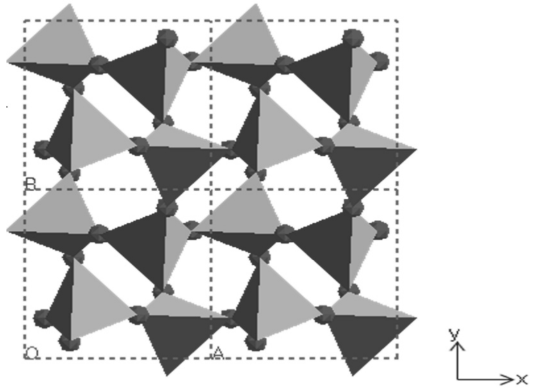


Fig. 5. Illustration of 2x2 α-cristobalite structure in projection x-y

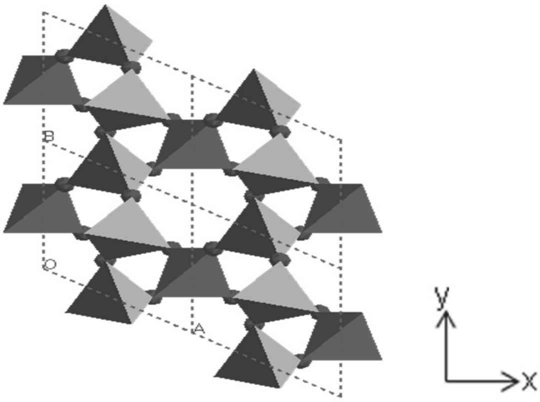


Fig. 6. Illustration of 2x2 α-quartz structure in projection x-y.

Zeolites are another important class of polyhedral framework nanostructures and several idealized zeolitic cage structures have been suggested to possess negative Poisson's ratios, predicted by Grima et al. 2000 using force-field molecular modelling simulations. Experimental confirmation of auxetic behaviour has subsequently been reported for Zeolite NAT (natrolite) by Sanchez-Valle et al. 2005 [48].

Apart from those naturally occurring inorganic materials, there are also organic materials with monoclinic crystal structure that have been very recently found to display auxetic properties. For example, crystalline cellulose I_β has recently been reported by Peura et al. in 2006 as having negative Poisson's ratios in the x-z plane for loading along the z-axis (cellulose chain direction), as characterized by x-ray diffraction. Nakamura and coworkers 2004 reported that cellulose II has auxetic behavior in the $(1\,\bar{1}\,0)$ plane for a uniaxial tensile load in the z direction (cellulose chain direction) [49].

Molecular level auxetics II: Synthesis of nanoauxetics

The re-entrant honeycomb structure is the first structure that has been identified as having a negative Poisson's ratio by deforming through flexure. This structure has been modelled analytically by Abdel-Sayed et al. and Gibson et al. [50].

Fig. 7(a) shows the deformation mechanism of the re-entrant honeycombs. For a re-entrant hexagonal geometry (Fig. 7(a)), under stretch in the y direction the cells elongate along the y-axis and open up in the x direction, leading to a negative Poisson's ratio.

Recent attention has focused on the theoretical design and synthesis of auxetic structures on a molecular level, and a number of organic auxetics have been proposed.

Generally, the design of molecular auxetics is being approached by first designing auxetic macro structures, and then downscaling these to the molecular or nano level [51,52]. Evans 1991 successfully employed such a downscaling technique to propose and demonstrate through modelling polyphenylacetylene single crystalline networks based on the re-entrant honeycomb structure having auxetic properties (See Fig. 7(b)). For example, such a structure was shown to be auxetic (Poisson's ratios as low as -0.94) by force-field type calculations [53]. Evans et al. have also proposed a simple 2 dimensional analytical model to predict the mechanical properties of their so-called (n,m)-flexyne/reflexyne (n, m is number of triple bond) (conventional/re-entrant hexagons) networks. Deformation was assumed to be the concurrent stretching, hinging and flexing of the 'arms' of the molecular network. The vertical and diagonal arms contain m and n acetylene links, respectively.

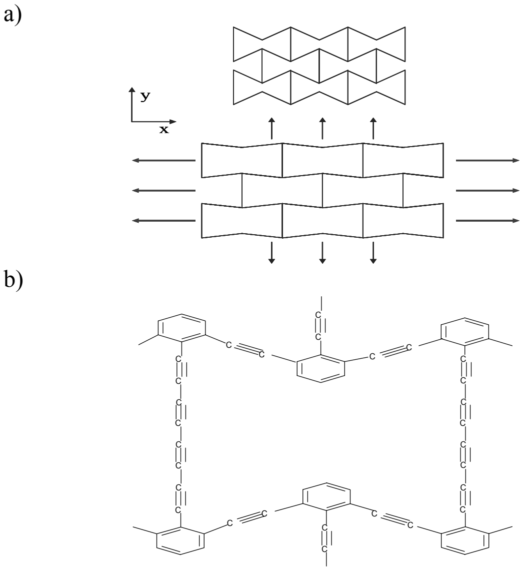
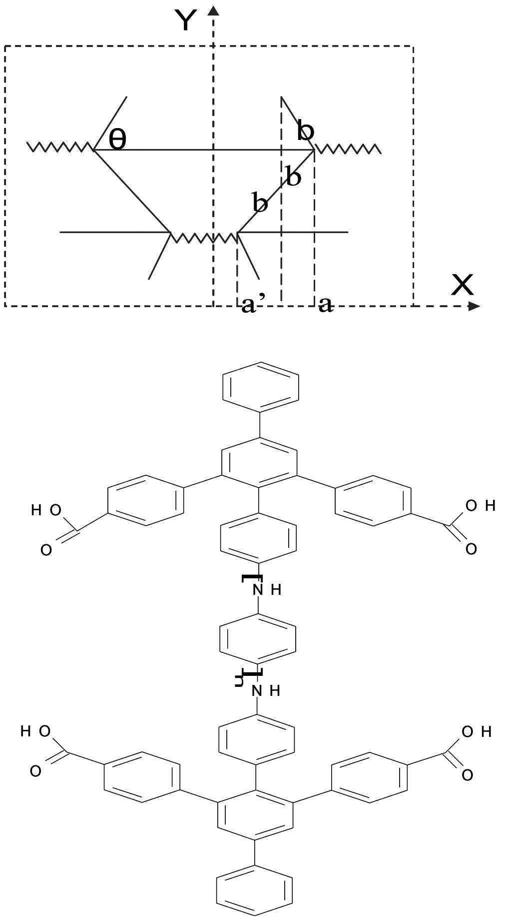


Fig. 7. (a) Two-dimensional deformation mechanisms in auxetic honeycomb structure, which are subjected to loading in the y-direction. (b) Auxetic molecular honeycomb network: (n,m)-reflexyne [1]

While the re-entrant honeycomb structure can in principle provide a polymer with the desired auxetic properties, it also would lead to a high melting temperature and poor tractability as a result of their highly cross-linked structure [54]. Consequently, these particular materials are not likely to be easily synthesised or processed [55]. Additionally, the auxetic behaviour only appears in one plane, and a bulk material consisting of randomly oriented crystallites may be not auxetic.

Wei 2005 developed the flexyne/reflexyne networks further and proposed self-assembled copolymers based on a hydrogen-bonded polymer network, shown in Figure 8. Alternating copolymers having a double-arrow-like "hard" block and a spring-like "soft" (-O-CH2-CH2-O- unit) segment have been designed which can in principle self-assemble into two-dimensional supramolecular networks [56].

This hypothetical polymer has the advantage of being more processible than the auxetic molecular network proposed by Evans et al, and the auxetic property has been predicted in Molecular Mechanics simulations, in the plane of the structure.

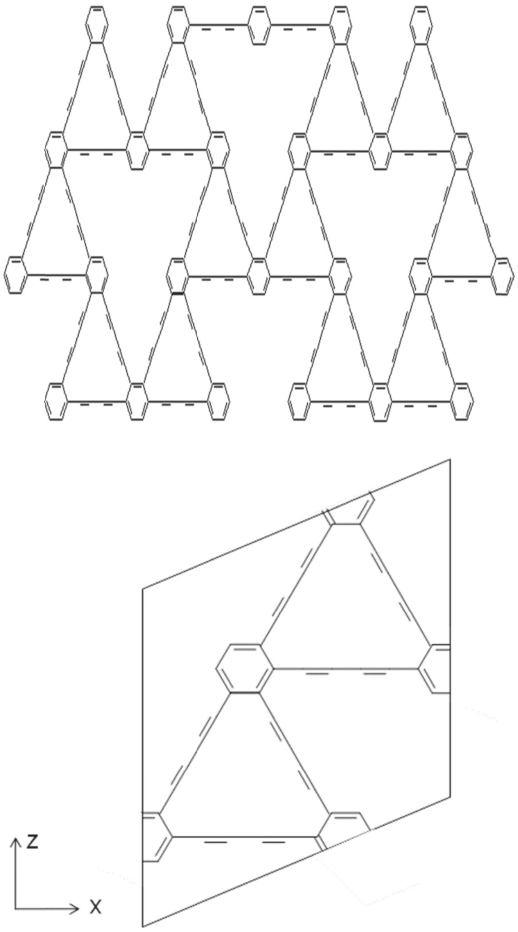


a)

b)

Fig. 8. (a) repeating unit of the self-assembly and the relationship among the variable *b, a, θ* and *a'*, where *a* the half-length of the backbone in the double-arrow and *b* the length of the arm, *θ* the angle between the arm and the backbone, and *a'* the half-length of the spring; (b) an example of double-arrow-like molecules [56]

Grima and Evans 2000 proposed a series of self expanding molecular networks (Fig. 9) also based on polyphenylacetylene arrows. They are described as parallel 'graphite-like' layers each containing a planar polyphenylacetylene infinite network (tessellation of a repeat unit having two equilateral molecular triangles at 60° to each other).



a)

b)

Fig. 9. (a) the structure of polytriangles-2-yne; (b) the minimum energy configurations of polytriangles-7-yne at σ_z=0.0 GPa [57]

Negative Poisson's ratios have been obtained in 3 directions using Mechanics simulations and maximum auxeticity is found in plane y-z (ν_{yz}=-0.97 and ν_{zy}=-0.96), and explained by cooperative rotation of the corner-sharing triangles. In the plane x-y and x-z, negative Poisson's ratios are due to a variation in the amount of interactions between the different layers. The 'rotating triangles' deformation mechasnism was also used to interpret the auxetic response of auxetic zeolites [57,58]. Other zeolites are predicted to achieve auxetic response through rotation of corner-sharing rigid 'square' molecular units [48].

Grima et al, 2005 have also proposed a range of auxetic networked polymers based on calix[4] arene molecular building blocks. These were produced by downscaling the folded macrostructure shown in Fig. 10, which is known to display auxetic behaviour. As with the polyphenylacetylene networks, the molecular network shown in Fig. 10(b) is not easily synthesised.

So far, one of the most simple and promising approaches towards a syntheticaly accessible molecular-level auxetic polymer is the liquid crystalline polymer concept, proposed and demonstrated by Griffin et al 1995; Griffin & Liu 1998 and shown in Fig. 11. The liquid crystalline polymers are designed based on site-connectivity driven rod reorientation in their main chain to achieve auxetic behaviour. The main chain consists of 'rigid' rod molecule interconnected by 'flexible' spacer molecule. Some rigid rods are attached at their ends to the flexible spacer molecular, while other rigid rods have lateral attachment. Under a tensile stress, full extension of the polymer main chain will force the laterally attached rods from a position roughly parallel to the tensile axis to a position normal to it and push the neighbouring chains further apart (See Fig. 11).

The main advantage of this polymer network is that in principle, as it is liquid crystalline, it should be much easier to align the chains, leading to more covalent than non-covalent bond force transfer [59]. Unfortunately, mechanical tests on these materials have as yet failed to measure a negative Poisson's ratio.

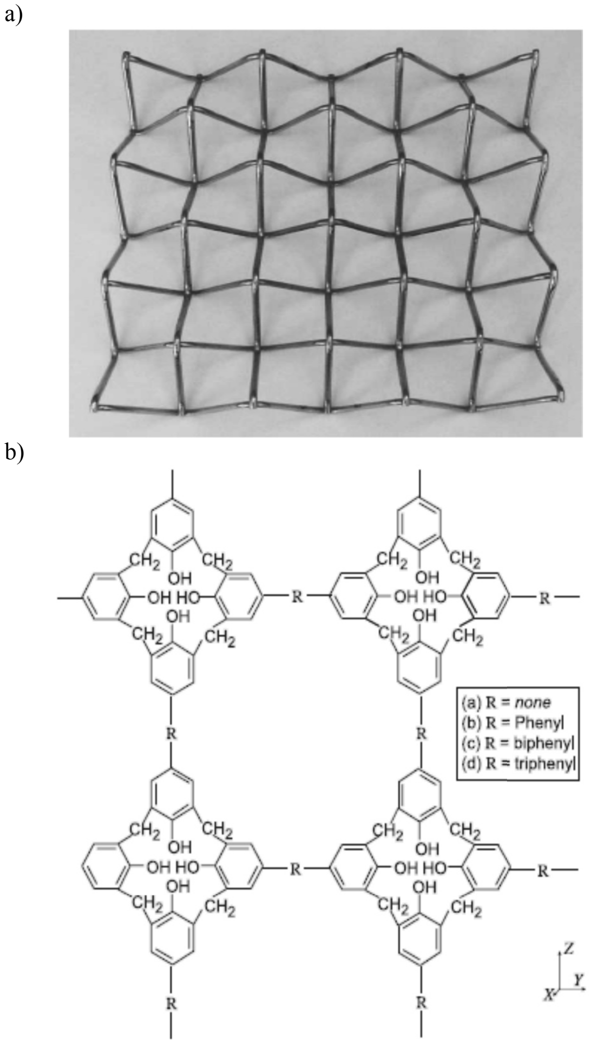


Fig. 10. (a) The auxetic macrostructure (30×30 cm) on which a theoretical molecular system is based. The macrostsructure is manufactured in steel and marketed as an 'egg rack'; (b) the molecular 'double calix[4]' network analogues [59]

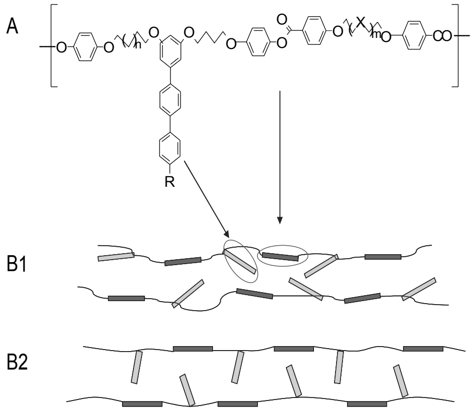


Fig. 11. (A) An example of one of the liquid crystalline polymers synthesised by Griffin and co-workers 2005 [58]: resorcinol-based terphenyl transverse rods (R=H, n-Pr, CN, CF_3, Ph) with possible spacers: methylene (X=CH_2), ethylene oxide (X=O), or siloxane (X=$Si(CH_3)_2O$). (B1 and B2) an illustration of the proposed mechanism resulting in auxetic behaviour: (B1) pre-align with surrounding liquid crystal field; and (B2) re-orient with a concomitant lateral expansion upon stretching

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