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An Alternative Explanation for the Negative Poisson's Ratios in Auxetic Foams

Joseph N. GRIMA*, Andrew ALDERSON[1] and Kenneth E. EVANS[2]

Faculty of Science, University of Malta, Msida MSD 06, Malta
[1]*Centre for Materials Research and Innovation, University of Bolton, Bolton, U.K.*
[2]*Department of Engineering, University of Exeter, Exeter, EX4 4QF, U.K.*

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Auxetic materials exhibit the unusual property of becoming fatter when stretched and thinner when compressed, in other words they exhibit a negative Poisson's ratio.[1)] A class of such materials which have attracted a lot of attention are auxetic foams[2–13)] which exhibit various enhanced physical characteristics.[6–9)] Foams with negative Poisson's ratios (see Fig. 1) were first manufactured by Lakes[2)] and can be produced from commercially available conventional foams through a process involving volumetric compression, heating beyond the polymer's softening temperature and then cooling whilst remaining under compression.[2–5)]

Various two-dimensional models which represent a cross-section of foams have been proposed in an attempt to relate the experimentally measured values of the Poisson's ratios to the microstructure of the foams. For example it has been proposed that conventional foams can be modelled using hexagonal[10,11)] and diamond-shaped[5)] honeycombs whilst the auxetic foams can be modelled through modified versions of these honeycombs.[5,10,11,14,15)]

In the case of the 2D hexagonal honeycomb model, the required structural modification for auxetic behaviour requires the junctions connecting the honeycomb cell walls ('ribs') to be transformed during processing from 'Y'-shaped joints to 'arrow head'-shaped joints.[10,11,14,15)] For the diamond honeycomb model, auxetic behaviour requires selective removal in a regular manner of ribs during the transformation process.[5)] One should note that these two modifications need not be simultaneously applied for a foam or honeycomb to become auxetic. For example, the creation of acute angles in the hexagonal honeycombs through the conversion of the 'Y'-shaped joints to 'arrow head'-shaped joints[10,11,14,15)] is enough to make the honeycombs auxetic without the need of altering the topology of the cells by the removal of ribs.

Although the above models probably play some role in modelling the auxetic behaviour in foams, and these models can reproduce the experimentally measured values of the Poisson's ratios, one may argue that there is not enough experimental evidence to justify the assumption that either of these are the main structural modifications which result in the observed auxetic effect. For example, whilst there is experimental evidence that there are 'broken ribs' on the surface of the auxetic foams [see Fig. 1(b)], it is still not clear whether 'broken ribs' are also present in the bulk of the foam material. Also, the requirement in the diamond honeycomb model for the removal of ribs in a regular fashion is not likely to occur in the existing foam conversion process. In the case of the hexagonal models there is no clear experimental evidence that a majority of the 'Y' shaped joints in the conventional foam are converted to the required 'arrow shaped' joints during the compression/heat treatment process. In fact, one may argue that it is unlikely that the majority of the changes in the foam manufacture process are concentrated at the joints of the foam as one usually observes that the ribs of open cell foams are slightly thicker in the proximity of the joints than at the centre of the ribs.

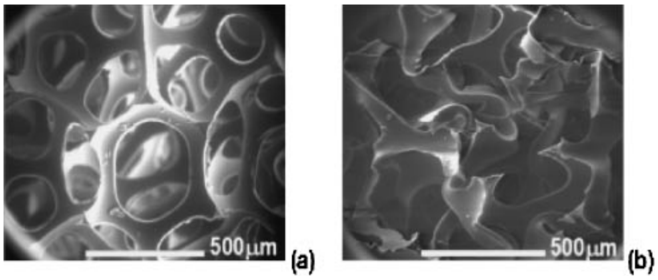

Fig. 1. SEM images of (a) conventional (non auxetic) open-cell polyurethane foams, and (b) auxetic open-cell polyurethane foam.

In view of this we propose a new model to explain the presence of negative Poisson's ratios in foams. This new model is based on the hypothesis that it is more likely that changes in the microstructure during the compression/heat treatment process will conserve the geometry at the joints (i.e., they do not become re-entrant) and the topology of the system (i.e., there are no rib breakages, as was the case in the re-entrant systems) and instead, the major deformations will occur along the length of the ribs which buckle (the foam is typically subjected to *ca.* 30% compressive strain along each axis[5)]). Figure 1(b) provides clear evidence of the presence of buckled ribs in the transformed auxetic foam microstructure. We also assume that the additional thickness in the proximity of the joints will make it possible for the joints to behave, to a first approximation, as 'rigid joints'. It is proposed that the rigid joints rotate relative to each other during the foam conversion process. The foam microstructure then 'freezes' in this much more compact form when the foam is cooled to below its softening temperature.

An illustration of this is given in Fig. 2 which shows how a conventional two-dimensional hexagonal honeycomb in Fig. 2(a) (which can be treated as a two-dimensional model for conventional foams) can be converted through the compression/heat treatment process into an auxetic form shown in Fig. 2(b). We propose that the 'rigid joints' behave like 'rigid triangles' [Fig. 2(a)] which, during the heating/compression process, rotate relative to each other to produce the more compact microstructure shown in Fig. 2(b). (This occurs though the formation of 'kinks' at the centre of the ribs which are the result of extensive buckling of the ribs in the compression/heat treatment process.) Uniaxial tensile loading of the idealised microstructure in Fig. 2(b) will cause a re-rotation of the triangles to generate the auxetic effect as illustrated in Fig. 2(c). (This corresponds to re-

*E-mail: joseph.grima@um.edu.mt

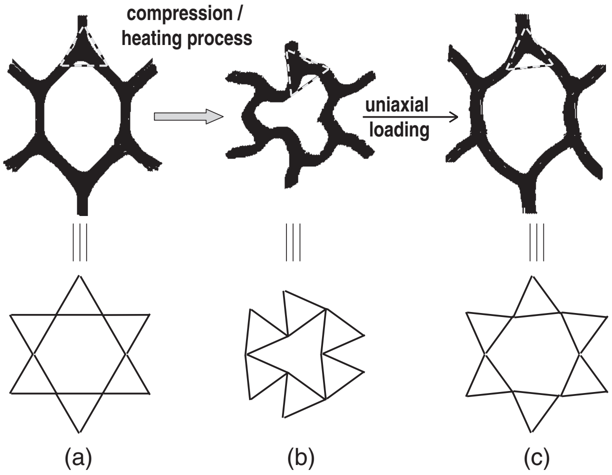

Fig. 2. The newly proposed model (top) and the idealised representation in terms of 'rotating triangles' (bottom): (a) The conventional 'hexagonal honeycomb model', (b) the compressed microstructure produced in the compression/heating process, and (c) the re-opening of the microstructure as a result of uniaxial loads. Note that in this newly proposed model the 'joints' of the honeycomb are behaving as rigid units, which rotate relative to each other through folding/unfolding of 'kinks' at the centre of the ribs. In an idealised scenario, the hexagonal honeycomb may be described in terms of 'rigid triangles' (which corresponds to the rigid joints) connected together at their vertices through hinges (which correspond to the 'kinks' at the centre of the ribs of the honeycomb).

unfolding of 'kinks' at the centre of the ribs and rotation of the 'rigid joints' which results in auxetic behaviour.)

It should be noted that similar 'rotation of rigid units' mechanisms have been found to play an important role in generating auxetic behaviour in various other classes of materials including zeolites,[16–19] nanostructured polymers,[17,20] silicates[17,21] and single crystalline materials belonging to the KH_2PO_4 (KDP) family.[22] It has been shown that for such mechanisms, the extent of auxeticity depends, amongst other things, on the particular shapes and the rigidity of the 'rigid units'.[15,23,24] In the idealised scenario presented here (Fig. 2) where the 'rigid units' are perfectly rigid equilateral triangles, the system would exhibit in-plane Poisson's ratios of -1. In practice, this value would be expected to be less negative since in reality the joints are unlikely to be perfectly rigid. Typically, structurally modified foams via the compression/heat treatment process have a value for the Poisson's ratio of ~ -0.7.[2]

One should note that uniaxial loading of the idealised 'rotating triangles' structure in Fig. 2(a) will not result in a negative Poisson's ratio as this is a fully extended structure. The structure in Fig. 2(a) will instead be constrained to deform through other modes of deformation (predominantly through flexure of the ribs[10,11]) which will result in conventional behaviour. This is consistent with the fact that the fully extended hexagonal honeycomb structure is suitable for modelling the behaviour of conventional foams.[5]

It is worthwhile to note that in reality, the 'rotation of rigid units' mechanisms operating in foams are likely to be much more complex, where for example, the junction units will be three dimensional and non-rigid to some degree. (Various three-dimensional models for auxetic foams involving re-entrant units have already been proposed, including models based on dodecahedron[12] and tetrakaidecahedron units,[13] and we envisage that a three-dimensional equivalent of our newly proposed two-dimensional 'rotation of rigid joints' model is also feasible.) We also envisage that in reality this new mechanism will work in parallel to other mechanisms such as the ones referred to earlier.[5,10–15] However, we consider the junction rotation mechanism is likely to be one of the predominant mechanisms for the production of the observed auxetic effect in structurally modified foams.

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