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Manufacturing, characteristics and applications of auxetic foams: A state-of-the-art review

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14 Abstract:

15 Auxetic foams counter-intuitively expand (shrink) under stretching (compression). These 16 foams can exhibit superior mechanical properties such as resistance to shear and indentation, 17 improved toughness and energy absorption (EA) under several types of loadings. Their unique 18 deformation mechanism and manufacturing process lead to special multiphysics properties such as 19 variable permeability, synclastic curvature and shape memory. Except for traditional energy 20 absorber stuff, the potential applications of auxetic foams have involved biomedicine, aerospace, 21 smart sensing, etc. However, most of the potential applications are restrained in the theoretical stage 22 due to complicated fabrication and a deficiency of stability. For removing the barrier for practical 23 application, a series of issues remain to be resolved, though the explorations of the improved 24 conversion methodologies and potential applications are fruitful in the past decades. We present 25 here a review article discussing the state-of-the-art for manufacturing, characterization and 26 applications of auxetic foams. We also provide a view of the existing challenges and possible future 27 research directions, aiming to state the perspective and inspire researchers to further develop the 28 field of auxetic foams.

### Keywords: auxetic, foam materials, smart materials, negative Poisson's ratio, protection equipment

#### 32 **1. Introduction**

The Poisson's ratio (denoted v for isotropic materials) is a parameter that defines the ratio between the lateral deformation of a body subjected to axial loading, i.e., between the transverse strain ( $\varepsilon_l$ ) and the longitudinal strain ( $\varepsilon_l$ ) with the minus sign (- $\varepsilon_l / \varepsilon_l$  [1]). According to the classical theory of elasticity, the Poisson's ratio for isotropic materials ranges from - 1 to 0.5 [2], so that a negative Poisson's ratio is allowable in linear elastic and thermodynamically correct materials. Most of normal materials become fat (thin) under compression (tension), and so exhibit a positive Poisson's ratio behaviour. In contrast, negative Poisson's ratio materials show a counter-intuitive 40 behaviour under axial loading - they become thin (fat) under compression (tension) (Figure. 1).



#### Conventional

Auxetic

41 42 Figure. 1. Shape changes under stretching of conventional foams (left) and auxetic foams (right). 43 The term "auxetic" was introduced by Evans [3] who adapted the ancient Greek word "auxetos" 44 (that which tends to increase) for negative Poisson's ratio materials. Iron pyrite monocrystals was 45 reported as possessing an auxetic behavior since 1882 [2], with a Poisson's ratio value of - 1/7 46 estimated by Love [4]. Natural auxetic materials that have been reported in open literature also include cow teat skin [5], cat skin [6], cancellous bone [7] and membranes found in the cytoskeleton 47 48 of red blood cells [8]. Wojciechowski [9] also demonstrated the existence of negative Poisson's 49 ratio in cyclic hexamers molecule assemblies subjected to critical pressure levels.

50 Foam materials possess many superior properties, such as improved performance under the 51 impact, lightweight, cost-effective, reusable, desirable acoustic capability, and remarkable chemical 52 and physical stability [10-14]. In this context, foam material has been widely used in daily life and 53 is considered a potential candidate for multifunctional engineering materials. On the other hand, 54 foam materials have high resilience and can bear large strain deformation. Such properties are 55 desirable to auxetic materials which need enough axial deformation for the showing of unique lateral deformation. Therefore, it could be expected that foam material would exhibit more remarkable and 56 57 functional properties if it be endowed with auxetic performance.

58 The first artificial auxetic open cell foam fabricated using a combination of volumetric 59 compression and thermoforming was reported by Lakes in 1987 [15]. During the following decades, increasing numbers of artificial auxetic materials have been developed at different scales [16] 60 61 (Figure. 2) and one typical methodology, pattern scale factor (PSF) methodology was proposed to 62 artificially design the auxetic unit cells with tunable mechanical performance [17, 18]. A good 63 portion of artificial auxetic materials is fabricated via textiles or 3D printing, whereas auxetic foams 64 can be produced by post-processing existing conventional off-the-shelf porous materials.





Figure. 2. Categories of auxetics at different scales (Reproduced from [19]).

Though several reviews on auxetic materials have been published in the past decades [16, 19-28], a more focused review on auxetic foams is still rare. Considerable progress in the fabrication, characterization and applications of auxetic foams has been reported in the past eight years since the publication of last review on auxetic foams [29]. In this paper, state-of-the-art review and the problems accompanied with possible solutions of current researches on the manufacture, characteristics and applications of auxetic foams have been made, aiming to inspire the peers and pave the way for further studies (**Figure. 3**).





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#### 77 2. Manufacture methodologies for auxetic foams

78 The principle for transforming traditional foams into auxetic form consists in producing a 79 stable compression that induces the lattice into a re-entrant shape. The volumetric compression can 80 be generated either by mechanical or via air pressure at first, and the foams ribs can then be softened 81 into shape by applying a specific heating profile or chemical solvent, followed by the re-entrant 82 shape fixed by the annealing and a cooling stage. Softening and cooling are however unnecessary 83 for metal foams (copper foam) (Figure. 4) [30-33] due to the high plasticity and the possibility of 84 creating the re-entrant shape by plastic deformations only. Methodologies for the conversion of 85 auxetic foams are fruitful and still on-going, current foam materials that can be converted into auxetics are listed in Table. 1. 86



- Figure. 4. Microstructures of copper foam subjected to volumetric compression ratios (VCR: Initial /
- Final Volume) of (a) 1, (b) 4.43 and (c) 4.94 respectively (Adapted from [33]).
- 90 **Table. 1.** Examples of several foam samples that have been managed to be converted into auxetics

material		pore diameter (mm)	density (kg/m³)	reference
polyester urethane foam	closed-cell	0.42 (60 ppi)	37.9	[37]
	reticulated	0.42 (60 ppi)	33.7	[37]
	open-cell	2.54 (10 ppi)	24.1	
		0.85 (30 ppi)	24.5	[37]
		0.42 (60 ppi)	21.7	
open-cell polyurethane foam		2.54 (10 ppi)	34	[70]
		1.27 (20 ppi)	30	[66]
		0.73-0.85 (30-35 ppi)	32	[113]
		0.73-0.85 (30-35 ppi)	27	[144]
		0.56 (45 ppi)	27	[70]
		0.45-0.49 (52-57 ppi)	27	[38]
		0.39 (65 ppi)	30	[66]
		0.25 (100 ppi)	30	[66]
closed-cell polyethylene foam		20 (expansion ratio)	not given	
		30 (expansion ratio)	not given	[53]
		45 (expansion ratio)	not given	
closed-cell polyvinyl chloride (PVC) foam		not given	52	[53]
open-cell copper foam		1	not given	[30]
room temperature vulcanizing elastomeric silicone foam		not given	150	[30]
closed-cell polymethacrylimide foam			52	
		not given	205	[44]
			301	
			59	
closed-cell polyethylene foam		not given	109	[44]
			158	

("ppi" means "pores per inch" - the most common unit for foam pore size).

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#### 93 2.1 Original and improved thermo-mechanical methodologies

94 In 1987, the first auxetic foam with Poisson's ratio value of - 0.7 was converted from 95 conventional commercial open-cell polyurethane (PU) foam by Lakes [15] through a thermo-96 mechanical (mechanical compression and thermoforming) methodology (Figure. 5a). Samples of 97 conventional foam were compressed along three orthogonal directions and placed in a mold at a 98 temperature slightly higher than the softening one of the foam samples (163°C to 171°C). The mold 99 was then cooled and the resulting foam showed stable buckled ribs and negative Poisson's ratio. 100 Uniaxial compression was also applied but could only enable the production of foams with a near 101 zero Poisson's ratio rather than a negative value [15]. However, further studies have shown that 102 foam samples that are sufficiently thin could provide a negative but inhomogeneous distributed 103 Poisson's ratio under only uniaxial compression [34, 35].

The thermo-mechanical methodology could also convert polyvinyl chloride (PVC) foams [36, 37], closed-cell PU [38] and silicone rubber foam [30] into auxetic foam. As for metal foam (copper foam), the heating stage is however not necessary because the conversion could happen under sequential plastic compression along three directions [30-33]. Friis *et al.* [30] produced auxetic PU foam by applying triaxial compression during the foaming process without the heating phase as well.

The toughest manufacturing limitations of the thermo-mechanical methodology are, especially
in manufacturing large foam samples, the sufficiency of long-term stability in mechanical properties,
risk of creasing surface and uneven heating and / or compression during manufacturing. To

112 overcome these drawbacks, several improvements based on the thermo-mechanical methodology 113 have been introduced (Figure. 5). A quasi tri-axial methodology (Figure. 5b) was described by 114 Mohsenizadeh et al. [39]. A novel mold was introduced to split the compression process into three 115 stages (x-compression, y-compression and z-compression), ensuring that foam samples are evenly 116 compressed in three directions. Foam samples fabricated using this methodology showed a better 117 homogeneity, rigidity and isotropic behaviour and effectively overcame the surface creasing problem. Duncan et al. [40] developed a novel cuboidal mold with through-thickness pins (Figure. 118 5c) to reduce folds on the foam surface and improve the uniformity of compression throughout the 119 120 sample. The use of pins to control the internal compression state of the foam, in addition to the 121 external compression applied by the mold, was first introduced by Sanami et al. in the production 122 of one-piece radially gradient "core-sheath" foams having distinctly different core and sheath region 123 pore structures, Poisson's ratios (including negative), and Young's moduli [41]. Subsequentially, 124 Duncan et al. [42] designed a novel mold with partly distributed through-thickness pins for fabricating gradient auxetic foam sheets. This mold was partitioned to generate different fabrication 125 126 and compression processes, which could significantly shorten the manufacturing time of different 127 types of foam samples. The work of Duncan et al. also highlighted the advantages brought by 128 through-thickness pins approach. Except for introducing novel molds to improve compression 129 quality, some additional processes are also effective to obtain auxetic foam samples with better performance in stability and mechanical properties. Chan et al. [38] proposed a multi-stage process 130 131 enabling conventional foam samples to be converted twice in different VCR (Figure. 5d) for a 132 gradual compression. This process can effectively avoid the creasing of the surfaces that occurs 133 during original one-stage compression and exhibits the potential benefit to manufacture large foam 134 samples. Bianchi [43, 44] applied a re-heating process (Figure. 5e) on the samples recovered from 135 auxetic foams. The stability of the auxeticity and the mechanical properties of the auxetic foam 136 samples could be significantly enhanced by re-manufacturing.



Figure. 5. Current methodologies based on the thermo-mechanical methodology of (a) original thermomechanical methodology [15], introducing of (b) quasi tri-axial mold [39] and (c) cuboidal mold with through-thickness pins [40] for uniform compression, (d) adopting a pre-auxetic process to realize multi-stage (gradual) compression [38], and (e) a double thermo-mechanical process to convert the recovered foam to obtain twice-processed auxetic foam samples with long-term shape stability [43, 44].

143 Two alternative compression techniques to produce the volumetric compression have been also elsewhere developed. The early study of Martz et al. [45] reported that closed-cell 144 145 polymethacrylimide (PMI) and low-density polyethylene (LDPE) foams could show an auxetic 146 behaviour under high external air pressure followed by heating process. Air pressure (or suction) 147 was also used in the "half mould" manufacturing process proposed by Bianchi et al. [46] to 148 manufacture auxetic foams samples with curved and arbitrary shapes. By using engineered vacuum, 149 Zhang et al. [47] fabricated foam samples having a stiffness 5 times larger than the stiffest auxetic 150 foam presented in open literature, meeting the requirements of both high stiffness and auxeticity. 151 Hydro-static pressure has been also applied by Najarian et al. [48] as another tool to compress the 152 foam samples.

# *2.2 Adopting of acetone, CO2, steam penetration and condensation (SPC) and 3D printer*

One of the earliest explanations of the auxetic behavior in negative Poisson's ratio foams is 155 156 based on the "missing rib" concept, i.e., the failure and breaking of ribs during the volumetric compression leading to changes of the original pore cell geometry. Smith et al. [49] therefore 157 158 predicted that foam ribs might be removed following a chemical process so the foam samples could 159 turn into auxetics. In 2009, Grima [50] realized the predicted conversion of auxetic foams by placing compressed foam samples within a container in acetone to soften foam ribs, rather than applying a 160 161 temperature profile for the annealing. Inspired by this novel softening process, an improved chemo-162 mechanical methodology (the Mechanic-Chemic-Thermal technique) (Figure. 6) was then developed by the Air Force Institute of Technology (AFIT) in Poland [51]. Compared with foams 163 164 fabricated using the original thermo-mechanical methodology [52], auxetic foams softened by 165 chemical process could obtain a lower Poisson's ratio without the risk of uneven heating, and this 166 approach is potential to produce large samples, however in considering of the safety hazard and environmental footprint issues, such chemical softening process is not recommended in further 167 168 researches.



169Final setting (h at 120°C)170Figure. 6. Flow chart depicting the four stages auxetic foam manufacturing process proposed by AFIT171(Adapted from [51]).

For developing a more environment-friendly fabrication process, Li [53] adopted  $CO_2$  as the main softening agent. Carbon dioxide could reduce the glass transition temperature of Styrene acrylonitrile copolymer (SAN) particles (copolymers in PU foams, play the key role in foam ribs softening) after a series of chemical reactions. Auxetic foams treated with carbon dioxide could be rapidly converted at a room temperature and exhibited a nearly constant value of negative Poisson's

177 ratio over a large strain range. Recently, basing on the function of CO<sub>2</sub> in the manufacturing, high-

performance closed-cell auxetic nylon elastomer (NE) foams with Poisson's ratio value of - 1.29 178 179 and superior mechanical properties have been fabricated by Fan et al. [54] using a green and 180 environment-friendly methodology named One-Pot CO<sub>2</sub> foaming process (Figure. 7a). Fan et al. have also remarked that this methodology could be also applied to other polymers (like ethylene-181 182 vinyl acetate (EVA) resins). SPC methodology (Figure. 7b) was described by Fan et al. [36], which enabled the development of auxetic closed-cell polyethylene (PE) and PVC foams. This 183 184 methodology could also be used for fabricating large auxetic foam samples as a related paper of 185 Duncan et al. [55] shows.



Figure. 7. Schematic drawings showing the experimental flow chart (left) and possible formation
mechanism (right) of a) One-Pot CO<sub>2</sub> foaming process and b) SPC process (Adapted from [54] and
[36]).

190 With the rapid development of 3D print technology, complicated 3D structures can be 191 fabricated with increasingly high precision in 3D printers. Except for traditional methodologies to 192 convert foam materials into auxetics, a novel concept of manufacturing auxetic foams by 3D print 193 technology was proposed by Critchley et al. [56], the foam samples they manufactured were 194 customized designed and fabricated without any random cell orientation, providing a new 195 methodology to generate foam structures with stability and homogeneity (Figure. 8). However, relatively high costs, scale limitation and the presence of defects still impede the broad application 196 197 of this technique. From this perspective, the rest of porous auxetic structures fabricated by 3D 198 printers could be defined as a novel foam material in a broad sense (Syntactic foams [28, 57-65]). 199 Such structures are potential alternatives as polymer foam materials to some applications such as 200 sensors or soft robots, providing stable and precise reactions under loadings.



**Figure. 8**. The unit cell (left) and the whole model (right) of auxetic foam fabricated by 3D print technology [56].

Although the methodologies mentioned above are feasible to convert conventional foams into auxetics, most of them are restricted in a small dimension. Large samples cannot be compressed and softened in an even manner, resulting in sometimes substandard performance. Many potential applications for which auxetic foams could be extremely advantageous to use (cushions and pads, for example) are still confined at the stage of design and prototyping, waiting for low-cost and efficient large scale manufacturing routes to be fully developed.

#### 210 2.3 Parametric manufacturing

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211 Several research teams have looked at identifying the most important manufacturing 212 parameters that improve the quality and performance of auxetic foams. In relation to the thermo-213 mechanical methodology, the debate about the best combinations of those parameters (temperature, 214 heating time and VCR) has started since the beginning of the original productions of auxetic foams. 215 The first negative Poisson's ratio polymer foam fabricated by Lakes [15] was subjected to VCR values from 1.4 to 4 and heating temperatures between 163°C and 171°C, heating time was however 216 217 unmentioned in his work. Friis et al. [30] indicated that 200°C may be a more suitable temperature 218 to soften the foam ribs for the tested PU foams of solid volume fraction 0.043. The heating time and 219 VCR adopted in the work are 7 min and 3.4, respectively. Wang et al. treated 40 100 ppi (pore 220 diameter of 0.25 mm) PU foam specimens (the density was 0.033 g/cm<sup>3</sup>) under different 221 combinations of manufacturing parameter to investigate the influence of the temperature and 222 heating time on the final value of the Poisson's ratio [66] (Figure. 9). In the work of Choi et al. [67], two types of open-cell PU foams (gray polyurethane-polyester foam:  $\rho/\rho_s = 0.03 \pm 10\%$ , L (length 223 224 of cell rib) =  $0.4 \pm 0.03$  mm and Scott industrial foam:  $\rho/\rho_s = 0.03 \pm 7\%$ , L =  $1.2 \pm 0.03$  mm) have 225 been produced by using different parametrical combinations. The lowest value of the Poisson's ratio 226 achieved in that work was - 0.7, for both kinds of foam, obtained from a VCR between 3.3 to 3.7 under a heating temperature of 170 °C for 17 min. Moreover, another paper of Choi' group shows 227

#### that the VCR should be smaller than 5 to avoid contact between ribs and adhesion [67].



Figure. 9. Poisson's ratio of auxetic foams fabricated from 100 ppi (pore diameter of 0.25 mm) PU foams with VCR of 2.5 and different combinations of temperature and heating time (Data replotted from [66]).

233 Martz et al. [45] noted that excessive temperature exposure (220 °C) could only make 234 transformed PMI closed-cell foams stiffer without auxeticity, the reason for this phenomenon may be the adhesion of melted foam walls. Duncan et al. [68] reported that a temperature higher than 235 200 °C would however induce a substantial mass loss of the open-cell PUR30FR foams, and a 236 237 combination of high temperature and long heating times could only make them exhibit auxeticity in tension. The optimal parametrical combination they provided are VCR = 3, 130 °C, 140 °C, and 238 239 160 °C for 180, 60, and 20 min, respectively, resulting in a minimum v = -0.2. As for heating time, 240 Chan and Evans [38] proposed that foam samples could not be "set" and were apt to recover to 241 their original shape (or be melted into a block of dense material) if the heating time was too short 242 (or too long in the case of melting). The critical temperatures of reticulated PU foams in three phases 243 (softening around 180 °C, liquefaction around 270 °C and polymer decomposition around 300 °C) were also noted in the paper. Temperatures about 5 - 20 °C lower than the softening temperature 244 245 were suggested advisable to maximize the stress relaxation and minimize the possible bonding 246 between cell ribs. Wang and Lakes [66] then proposed a series of optimal combinations of 247 temperature and heating times for open-cell PU foam samples with different cell sizes (25 ppi, 65 ppi and 100 ppi, i.e., pore diameter of 1 mm, 0.39 mm and 0.25 mm, respectively). All samples were 248 249 produced with the same VCR of 2.5. Detailed parameters and results are presented in Figure. 10. 250 The first design of experiment (DoE) work related to the manufacturing of open cell PU auxetic foams was carried out by Bianchi et al. [69]. The Authors of that work carried a systematic campaign 251 252 of quasi-static cyclic tension/compression tests on 80 different cylindrical foam specimens with

VCRs ranging between 5 and 19, two temperatures (135 °C and 150 °C), two heating times (12 min 253 and 15 min) and also two different cooling methodologies (water and air ventilation). The final 254 conclusions of the DoE showed that the VCR is the most statistically important manufacturing, 255 while no significant statistical correlation could be identified with the temperature, heating times 256 257 provided and the cooling methodologies. Critchley et al. [70] recently provided an updated 258 statistical approach for the optimization of the manufacturing parameters to improve the efficiency 259 of the production. An optimal combination of VCR, porosity and heating time was recommended for a conversion temperature of 200 °C, detailed parameters are also shown in Figure. 10. In view 260 261 of practical applications, especially for applications in EA, the final density is however indicated as a critical parameter [69, 71, 72]. A lower VCR appears to be preferable to maximize EA under quasi-262 263 static compression [73]. A VCR value of 2.2 was recommended as the best for PU foams used in 264 seat cushions [71].

- 265 To determine the effect of the material plasticity on energy dissipation of auxetic metal foams,
- 266 numerical impact tests on foam finite element (FE) models have been carried out by Kumar *et al.*
- 267 [74]. The plastic Poisson's ratio (the Poisson's ratio on the plastic stage  $v_p$  should be close to zero 268 to obtain the maximum energy dissipation.



Temperature (°C)
Figure. 10. Summary of the recommended parametric combinations based on experiments in the
literature of heating temperature, VCR and heating time (indicated by the number in blocks, measured in
minutes) for different foam samples (indicated by the color of blocks) (Data obtained from [66-68, 70,
71]).

PU foam samples were compressed using hydro-static pressure before being placed into the mold in the work of Najarian *et al.* [48], so the value of the pressure is also a critical parameter during in the conversion process in the work. In order to obtain foam samples with the best auxeticity and stiffness, a grey relational analysis (GRA) was proposed in the paper to control the influential parameters *viz.* temperature, pressure and time. The optimal combination of parameters involved the values of 140 °C for the annealing temperature, 40 bar for pressure and 20 min for heating time. Duncan *et al.* [55] recently investigated the effect of steam conversion on the cellular structure, Young's modulus and negative Poisson's ratio of closed-cell LDPE foams. The formation of cells with re-entrant shape tended to increase with the duration of steam conversion. Surprisingly, large foam samples in the fabrication shrank more evenly than small ones, indicating this methodology is a feasible way to fabricate large foam specimens.

Though lots of investigations have been conducted and the conclusions include detailed parametric combinations to manufacture optimal auxetic foams, the recommended combinations are regarded as reference to further experiments due to the variation of foam samples (such as chemical constituents and sample size) and randomness of manufacturing condition (such as room temperature for cooling). The size of foam samples, in fact, is of significance in the manufacturing consisting in heating process and should be taken into further consideration.

#### **3. Micro-structure and deformation mechanisms in auxetic foams**

Solid understanding of micro-structure and deformation mechanism in auxetic foams is not only the foundation to explore efficient manufacturing methodologies, but also desirable to create valid foam models using in numerical analysis and finite element modeling (FEA). Such models could also be utilized for inspiring auxetic structural design.

#### 296 3.1 Re-entrant polyhedron models of auxetic foams

297 The first step in developing micromechanical models for these foams is to define the geometry 298 of the original and converted foam cells. The internal pressure and squeezing of neighbor cells or 299 surfaces during the foaming stage generate cells with multiple polyhedron shapes. Most of the 300 earliest studies on foam structures have been focused on simplified topological representations 301 involving faceted polyhedrons (Figure. 11). Lakes [15] is the first to present stereo photographs and 302 an idealized re-entrant unit cell structure for the auxetic foams, in which the ribs were bent and 303 protruded into the cells: an axial tensile load would cause the cell to unfold and expand laterally. 304 Later, Friis [30] developed the topology of the model further by using the Kelvin minimum area 305 tetrakaidecahedron consisting in a convex shape with eight curved hexagonal faces and six curved 306 square faces. However, the description of how conventional foam deforms was absent. Wei [75] 307 extended the lattice formulation of Warren and Kraynik [76] to represent polymeric networks via 308 three arms of equal length and one backbone all connected to one junction. The model could be 309 applied to both 2D and 3D cases. Choi et al. [77] proposed a novel 3D tetrakaidecahedron model companied by a description of how it could assume a re-entrant structure. Doyoyo et al. [78] 310 311 presented a 3D auxetic structural lattice and verified that it could predict some mechanical properties 312 of previously published auxetic foam materials. Cham et al. [79] argued that the tetrakaidecahedron 313 model is not adequate to consider anisotropic foams with a preferential rise direction during foaming 314 or thermoforming, and therefore introduced a more generally accepted dodecahedron model.



Figure. 11. 3D polyhedral models of auxetic foams proposed in literature: (a) Idealized re-entrant
unit cell structure, original foam structure is not given in the paper [18,78], (b) Kelvin minimum area
tetrakaidecahedron model [30,77] and (c) Dodecahedron model [79].

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#### 319 3.2 2D models of auxetic foams

320 Various 2D models have also been developed to provide a more simple explanation of potential 321 deformation mechanisms occurring inside auxetic foams when subjected to loading (Figure. 12). 322 The 2D lattices derived from 3D configurations could be also helpful to inspire the design of general 323 2D auxetic materials. Early studies considered the auxetic structure of the foam as derived from the 324 hexagonal configuration lattice [42, 80, 81] and modified those centrosymmetric topologies into 325 auxetic ones [42, 80-83]. In those models the auxetic behaviour requires the deformation of the foam ribs from "Y" shape to "arrow-head" joints (i.e., the re-entrant structure). Another 2D model named 326 327 "missing rib" model was proposed by Smith et al. [49]. This model could explain the auxeticity and 328 the strain-dependent Poission's ratio behaviour by further fitting some of its parameters to 329 experimental data. Further modifications of the missing rib model were introduced by Gaspar et al. 330 [84] and Lim et al. [85], with improved accuracy in terms of the prediction of the Poisson's ratio of 331 the auxetic foams. However, Gaspar et al. [86] also noted that missing ribs are actually existing in 332 real auxetic foams, but their fraction over the total of the foam ribs is quite small and the dominant 333 deformation within the auxetic cells is still the bending of straight ribs.

334 In 2005, Grima [87] proposed a system based on rotating components linked by rigid or flexible

- 335 joints / ribs to explain the deformation mechanism in auxetic foams when compressed. The rigid joints inside the foams could be simplified into triangles [87-89]. Pozniak et al. [90] have developed 336 337 two 2D models of auxetic foams (Y and  $\triangle$  models). These two simple disordered structures could 338 predict well the deformation of foams under compression. An extended model was proposed by 339 Chetcuti et al. [89] that took into account the amount of material at the joints and the deformation of the joints themselves, this model allowed a set of more plausible predictions of the Poisson's 340 341 ratio values. Similar disordered structure for simulating auxetic foams is the 2D random cellular 342 solid model proposed by Li et al. [91]. Regular 2D Voronoi model was given a irregularity of 0.45 343 and then modified by a process of merging and removing to correspond with the geometric shape 344 of experimental auxetic foams. Alderson et al. [92] proposed three 2D topologies based on the 345 observation of micrographs of auxetic foams to illustrate the effects of uni-axial, bi-axial and tri-346 axial compression on the pore structure. Simular works have been carried out by Bianchi et al. [46] 347 for invastigating the deformation mechanism of converted foam samples fabricated by "half mould" 348 process.
- 349 Similarly to 3D models, the 2D models of auxetic foams allow to predict the auxeticity and 350 the mechanical of the porous materials. The deformation mechanisms predicted by these models
- 351 could also inspire future works in designing novel auxetic structures.



Figure. 12. 2D models of auxetic foams proposed in literature: (a) the transformation of "Y" shape joints to "arrow head" joints [43,80-83], (b) chiral structure produced by "Missing ribs"(broken ribs caused by compression, melt ribs caused by heating process or removed ribs caused by acetone) [48], (c) modified re-entrant "missing rib" model based on hexagonal foam cells [85], (d) regarding the joints as triangle shape, and the deformation based on the rotating of joints which resulting the re-entrance of foam ribs [87-89], and disordered foam structures based on (e) rotational joints model [90] and f ) 2D Voronoi model [91].

#### 360 3.3 Microphotographs and FE models of auxetic foams

The use of various imaging technologies such as digital image correlation (DIC), X-ray scattering, computed tomographic (CT) and scanning electron microscope (SEM) allows to identify the internal structure and deformation mechanism of auxetic foams (**Figure. 13**). Alderson *et al.* [34] observed that the auxetic behaviour of auxetic thin flat pads fabricated by uni-axial compression was due to a through-the-thickness crumpling of the cells, and evidence for the existence of the equivalent of rotation joints has been provided by X-ray microtomography [93, 94]. SEM has also allowed to observe bending, stretching, hinging and obvious rotations of elements of the foam pores during compression [73], indicating the presence of an heterogeneous deformation during the compression of foams. Similar studies based on CT techniques have been carried out by Elliott *et al.* [95], who also observed the presence of bending and entirely collapsed layers of cells and struts impinging on each other during compression.

SAN particles (mentioned in 2.2, the utilizing of CO<sub>2</sub>) were observed in PU foams by Li *et al.*[96], and this nanoparticle compound provided a likely explanation of the annealing temperature
mechanism within the fabrication of the foams. The finding of the consisting of SAN copolymers
also promotes further invastigations on auxetic foam manufacturing modyfying.

The imaging technologies mentioned above have also allowed to identify strong local heterogeneities of open-cell PU foams [85, 86, 97, 98], anisotropic behaviour in closed-cell PU foams [39, 97, 99] and open-cell PU foams [46, 47], difference in distribution of reduction in size of cells in the closed-cell PE foam [36] and defects of the foams fabricated using 3D print [56]. Recently, Pashineet *et al.* [100] interpreted the deformation of auxetic foams as a directed aging process, under which foam samples develop along an expected direction at larger scale like network bonds, rather than at microscopic one.



## **Microphotographs**

383 384

Figure. 13. Images of the microstructure of auxetic foams in literature: (a) Observations of
bending, stretching, hinging and obvious rotating during compression [52,55], (b) SAN particles which
provided a likely explanation of the annealing temperature mechanism within the fabrication of the
foams [96], (c) anisotropic behavior in auxetic foams [45,47], (d) local heterogeneities in auxetic foams
[108], (e) difference in shrank distribution in the closed-cell PE foam during the steam treatment [36]
and (f) defections of the foams fabricated by 3D print technology [56].

390 The imaging technologies allow the identification of foam structures and their deformation 391 mechanisms. Micrographs and 3D images obtained by using CT could also be used to develop solid

models of the foam skeletons and microstructures, and be transferred to the finite element analysis 392 (FEA) software for simulating the mechanics and deformation mechanisms of these foams at much 393 higher fidelity (Figure. 14). Two 3D region of interest models were crated and transformed into FE 394 395 models by McDonald et al. [93]. The local rib interactions and globle dispplacement of foam 396 samples can be faithfully represented by followed FEA process, providing confirmation to the 397 mechanisms observed in the experiments and greater details occurred in foam samples under applied 398 loadings. The above-mentioned 2D auxetic models that proposed by Pozniak et al. [90] and Li et al. [91] can also be built in FEA softwares. Pozniak et al. proposed two approaches for simulating the 399 400 structures they proposed : the plane stress based and the beam based approach. These two approaches are valid to simulate proposed structures, however, in consideration of both calculation 401 402 speed and simulating accuracy, the latter approach was recommended as the better one. As for the 403 works of Li *et al.*, different area compression ratios  $A_r$  was adopted in structural design to correspond 404 with the different VCRs that adopted in actual copper foams according to ref.[33]. The excellent agreement achieved between FEA and experimental observations shows that these irregular 405 406 structures can highly simulate the deformation of actual auxetic foam samples. Some mechanical 407 properties such as the Poisson's ratio behavior and EA capacity can also be predicted to some extent 408 by similation results of these structures. In order to investigate the blast resistance of auxetic 409 structures, Li et al. [101] built a series of 3D isotropic auxetic foam core in FEA software by using 410 random Voronoi structures. The Poisson's ratio varies from positive to negative by changing the 411 randomness and tri-axial compression ratio of the structures, providing a feasible methodology to 412 build auxetic FE models.

#### **Finite element models**



413
414
Figure. 14. FE models of auxetic foams in literature: (a) Two different FE model versions (plane
415 stress based and beam based) for the proposed disordered auxetic structures [90], (b) 2D random
416 cellular solid models based on a modified Voronoi tessellation technique for simulating the
417 compression of copper foam [91], (c) microstructurally faithful FE models created from the
418 reconstructed 3D tomography data [93], and (d) 3D isotropic auxetic foam core for blast resistance in
419 FEA software by using random Voronoi structures [101], the structures can be generated by using
420 different randomness (upper black ones) and tri-axial compression ratio (lower green ones).

#### 421 **4. Mechanical properties of auxetic foams**

The most particular property of auxetic foams is the possession of negative Poisson's ratio v, which is directly related, according to the classical theory of elasticity [102], to the Young's modulus E, the bulk modulus K (or resistance to changes to volume due to the application of a hydrostatic pressure) and the shear modulus G (or resistance to shear surface forces) in an isotropic material. The variation of the Poisson's ratio can significantly affect the mechanical behavior of the material. In an isotropic material with a constant E, if v approaches - 1, the shear modulus G theoretically approaches to infinity, i.e., such material could be easily compressed but hard to shear.

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

430 
$$K = \frac{E}{3(1-2\nu)}$$
 (2)

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

432

$$\nu = \frac{3K - 2G}{2(3K + G)}\tag{4}$$

433 Though auxetic foams are highly anisotropic [15, 19, 97, 99, 103-105] and other relations 434 between engineering constants need to be applied within the framework of classical elasticity to 435 predict the behavior of anisotropic auxetics [106-108], auxetic foams show indeed better indentation 436 shear resistance though the Young's modulus is smaller (lower E) than conventional ones [19, 31, 437 38, 45, 83, 108, 109]. The shear modulus in quasi-isotropic auxetic open cell foams can be well 438 approximated using the formulas related to classical isotropic elasticity [110], but the shear stiffness 439 of transverse isotropic auxetic foams has a different behavior [111]. The shear moduli vary between 440 10 - 38 kPa measured with a pure shear test [110] and 30 - 60 kPa via 3 point / 4 point bending tests 441 [111], with the latter measurements taken at higher strain rates.



442 443

Figure. 15. Synclastic (dome-shaped) curvature behavior of materials with a negative Poisson's ratio,
and drawing pictures present the ergonomic of this behavior (Images courtesy Andrew Alderson).
In terms of foam applications, the mechanical properties of stiffness (deformation resistance),
hardness (indentation resistance) and toughness (fracture resistance) are very important. Auxetic
foams also possess peculiar characteristics like the synclastic (dome-shaped) curvature (Figure. 15)
[15, 19], reduction of vibration transmissibility (Figure. 16) [112], acoustic absorption (Figure. 17)
[108, 112-118] and variable permeability (Figure. 18) [82, 119] Auxetic foams could also be made

- 450 reversible, which means that the foams can be converted to their original state by a reforming
- 451 process, i.e., they exhibit shape memory. (Figure. 19) [36, 43, 44, 54, 67, 74, 120, 121].





453

454

455

auxetic foams between 49 and 150 Hz at low amplitude vibrations via electrodynamic shaker excitations (Data replotted from [112]).

Figure. 16. Comparison between the vibration transmissibility (|T|) of conventional and



457 **Figure. 17**. Comparison of the frequency-dependent acoustic absorption between

458 conventional and auxetic foams measured via acoustic impedance tube (Data replotted from

459

456



[112]).

460
461 Figure. 18. Illustration of permeability tests with glass beads with auxetic foams, the foam
462 pores can be enlarged by stretching (Images from [119]).



463 464

Figure. 19. Shape memory behavior of auxetic PE foam (Adopted from [36]).

#### 465 4.1 Stiffness

466 The stiffness k is a parameter used to assess the deformation resistance offered by an elastic 467 body:

468

$$k = \frac{F}{\Delta l} \tag{5}$$

469 Where *F* is the force applied on the body and  $\Delta l$  is the resulting deformation of the body. The 470 stiffness and its evolution could significantly affect the EA and peak force under specific sets of 471 loadings.

The stiffness of auxetic foams is in general lower than their conventional counterpart at lower compression strains (**Figure. 20**) [30]. Chan and Evans [122] thought conventional foams need to overcome the stiffness of their hexagonal pristine structure before the ribs become bent, whereas the ribs of auxetic foams are already bent when testing. Such phenomenon was also found by Bhullar [123], and the paper proposed that the lower stiffness at small strains could be beneficial to biomedical implants.



478 Compressive strain
479 Figure. 20. Initial part of compressive strain-stress curves of auxetic foam and conventional foams
480 (Data replotted from [30]).

481 As the compression increases, auxetic foams shrink laterally due to their unique re-entrant 482 structure, making the density and stiffness increase significantly. Such phenomenon can be seen in 483 other auxetic structures and nowadays the stiffness control [124] during the loading process has 484 become a research point. On the contrary, conventional ones expand laterally and the density and stiffness in general feature more limited changes. Auxetic foams will therefore tend to be denser and 485 486 stiffer than conventional ones at higher compression strains. A lower stiffness of auxetic foams could 487 only be observed in a very small strain and hardly exists in samples having small scale pores. Foam 488 materials are widely used to undergo large deformation, so the lower stiffness in the very beginning 489 part could be negligible in most applications, although stiffness at small strain is also a parameter 490 that affect the static indentation of foams and the comfort provided in cushioning. The stiffness of 491 auxetic foams is sensitive to strain rate: the higher the strain rate be applied, the stronger the 492 stiffening effect [20, 125] (Figure. 21).



493 Compressive strain
 494 Figure. 21. Compressive strain-stress curves of auxetic PU foam at various strain rates (Data replotted
 495 from [20]).

#### 496 4.2 Hardness

497 The hardness of foam materials is one of the representative parameters to evaluate their 498 indentation resistance. The increased hardness of auxetic foams could be also predicted from the 499 perspective of the classical elasticity theory, the relationship of hardness (H) and Poisson's ratio ( $\nu$ ) 500 is given by:

501 
$$H \propto = \left[\frac{E}{(1-\nu^2)}\right]^{\gamma} \tag{6}$$

502 Where *E* is the Young's modulus,  $\gamma$  is 1 under uniform load or 2/3 under concentrated load. 503 When the  $\nu$  approaches -1, the stiffness approaches to infinity.

Superior hardness of structures with a negative Poisson's ratio was reported in previous experiments [126-128]. Auxetic materials feature a unique type of deformation under indentation: the material around the loading area will tend to concentrate towards the impact point, causing a significant increase of local hardness. On the contrary, materials with positive Poisson's ratio would expand outwards from the impact point in similar loading conditions, making indentation easier (**Figure. 22**). This indentation resistance behavior has also been experimentally observed in auxetic foams [45, 47, 117, 122, 129-135].



511
512 Figure. 22. Inner reactions under indentation of conventional (left) and auxetic foam (right).
513 Lisiecki *et al.* [51] argued that auxetic foams produced via the mechanic-chemic-thermal

514 process are not suitable for impact mitigation due to their tendency of becoming denser under 515 indentation or compression. Those authors performed quasistatic and drop tests, and indicated the 516 higher deceleration and smaller impact hammer displacements at an equivalent static pressure of

517 12.3 kPa as evidence for their conclusions (Figure. 23).



518 519 Figure. 23. Time histories of the hammer displacement (top) and acceleration (low) on auxetic and 520 conventional foams for an equivalent static pressure of 19.6 kPa (Data replotted from [51]). 521 Soft foams may however bottom out under indentation [112, 136], slightly higher stiffness of 522 auxetic foams may therefore provide more comfort as a cushion. Lakes and Lowe [71] noted that 523 the volumetric compression ratio (or the final density) is critical to obtain an optimal performance 524 under indentation in auxetic foams. Measurements of pressure distributions on PU foam cushions 525 were performed by those Authors, and the results showed that the re-entrant foam at densities between 0.032 and 0.064 g/cm<sup>3</sup> performed better (i.e., lower maximum seating pressure) than 526 conventional foam samples at comparable density. The auxetic foam material also wraps around the 527 528 indenter under indentation, which provides a more uniform stress distribution and a reduced 529 pressure concentration [71, 136, 137]. Further studies of Allen et al. [137] and Duncan et al. [138] 530 showed that the peak accelerations (synchronized with the maximum deformation) of flat indentors 531 used in drop tests on thermoformed-mechanical auxetic foams are smaller than in conventional ones 532 under low-kinetic energy impact conditions. This indicated that those auxetic foams are promising 533 for applications like cushions and pads (Figure. 24).



534Time (ms)535Figure. 24. Time histories of the acceleration for the flat indenter used in drop tests on R45FR foam at536(a) 2 J and (b) 4 J of kinetic energy (Data replotted from [137]).

#### 537 **4.3 Toughness**

Toughness indicates the ability of a material to resist fracture, and it is also a parameter adopted in crashworthiness designs [113, 139-141]. Toughness is in general critical for materials used in packaging and protection and it is measured by the area underneath the load-displacement or strainstress curve. That area is also the value EA, while the specific energy absorption (SEA) indicates the energy absorbed per unit mass of the structure. These two parameters could be calculated as follows [142, 143]:

$$EA(d) = \int_0^d F(x) \, dx \tag{7}$$

545

544

$$SEA(d) = \frac{EA(d)}{m}$$
(8)

546 In those equations d is the crushing distance, F is the crushing force and m is the mass of the 547 structure.

The parameters EA and SEA can be determined from quasi-static mechanical tests. Under quasi-static loading, auxetic foams absorb more energy than their conventional counterparts [40, 51, 54, 69, 101, 131, 132, 144-148]. A comparison of compression properties between auxetic foams

and PPR (Positive Poisson's ratio) foams in ref. [54] is presented below as an example in Figure.

552

25.



553 Strain(%)
554 Figure. 25. Compression of auxetic and the PPR foams: (a) compressive stress-strain curve, (b) EA
555 values for the auxetic and the PPR foams and (c) SEA for the auxetic and the PPR foams [54].

Mohsenizadeh and his group tested a series of auxetic foam-filled square tubes under both quasi-static and crushing tests with loading velocity of 1.5 m/s [139-141]. The EA value of the tube increases as the auxeticity of foam filler increases. The works of Scarpa *et al.* [80, 113] recorded the time histories of load and displacement in auxetic and conventional foams under high strain rate compression (**Figure. 26**). The time histories related to the auxetic foam demonstrated a better consistency, indicating a significantly improved resilience of the auxetic foam compared to the pristine conventional foam samples.



563 Time (sec)
 564 Figure. 26. Time histories of displacements and loads for (a) auxetic and (b) conventional foams under
 565 dynamic crushing loading conditions (cam plastometer) at 15 s<sup>-1</sup>. The full curve represents the force
 566 history and the broken curve represents the displacement history (Data replotted from [113]).

A series of fatigue tests were carried out by Bezazi and Scarpa [131, 149] to investigate the 567 toughness, stiffness loss and damping capacity of auxetic foams under cyclic loading (Figure. 27). 568 In those studies, the maximum load F was recorded as a function of the number of cycles N and 569 570 normalized by the maximum load  $F_{\theta}$  obtained during the first cycle. The loading level r was defined 571 as the ratio between the maximum displacement at a particular level to the displacement at failure. 572 A larger energy dissipation was observed in the displacement-load curve of the first cycle and the auxetic samples show a lower stiffness degradation (rigidity loss) ( $F/F_0$ ) after a large number of 573 574 cycles compared to the parent conventional ones.



at different loading levels of (a) r = 0.95 and (b) r = 0.75; (c) Stiffness degradation versus number of cycles for the two types of foams at different loading levels: r = 0.95; r = 0.725 (Data replotted from 579 [131]).

Another less documented but important parameter is the fracture toughness evaluated by Choi and Lakes [31] in auxetic copper foams, with significant improvements compared to the conventional copper foam version. Although those enhanced properties in fracture are promising in terms of the load-bearing applications, to the best knowledge of the authors of this review, no other work has focused on the fracture toughness of auxetics yet.

#### 585 **5. Applications of auxetic foams**

575 576

586 Auxetic foams are in essence transformed from traditional porous materials. The 587 transformation leads to a set of very desirable properties that conventional foams do not possess. It 588 is therefore reasonable to consider auxetic foams either as a replacement of conventional porous 589 materials, or as the basis of completely new designs to exploit their ad-hoc properties. Foam 590 materials are commonly used as fillers for cushion and auxetic foams, compared to conventional 591 ones, may further improve comfort and reduce the risk of pressure sores because of their EA [51] and pressure reduction [71, 136, 150]. These superior properties are also highly attractive for 592 protective equipment in sports applications such as pads or mats [51, 71, 136], gloves [144], helmets 593 594 [132, 151], spots shoes [132] and snow-sport safety devices (body armour and crash barrier) [145].

595 Several thin-walled tubes with auxetic foam core have been designed by Mohsenizadeh et al. 596 [140, 141, 152]. Compared to the control group made with conventional foam, the compression 597 induced shrinkage (density increasing) of the auxetic foam core provides enhanced EA and 598 crashworthiness (**Figure. 28**). Recently, Zhang et al. proposed a series of auxetic tubes with superior 599 mechanical properties [153, 154], compared with the ordinary tubes as the above-mentioned ones, such novel auxetic tubes could further enhance the mechanical performance of auxetic foam-filled

601 tubular structures.

602 603



Figure. 28. SEA and sections of foam-filled tubes (Adapted from [153]).

604 Sandwich composite beams with auxetic foam core could bear large loading (high shear stress) 605 under large deformations [111] and possess significant mechanical impedance with an interesting 606 zero stiffness behavior [155] (Figure. 29). Depending on the type of application and loading, auxetic foams could be used to tailor the performance and the overall weight of the structural sandwich 607 608 panel in which they are embedded. Zahra et al. [156] have described the manufacturing and 609 characterization of cementitious polymer mortar-auxetic foam composites and showed that the 610 embedding of the auxetic foam could avoid delamination and brittleness of the cement (Figure. 30). The unique deformation under compression (material concentration at compression area (Figure. 611 612 22)) endows auxetic foam with enhanced resistance for fracture. Such outstanding performances are 613 potential in the engineering field. Except for axial compression, it is also worth researching to test 614 auxetic structures under complicated loading conditions.

Recent progress in seismic metamaterial was reported by Huang *et al.* [157], a series of numerical simulations certified the positive function of auxetic foams as the shell to attenuate Lamb wave, realizing the generating of ultra-low frequency bandgap (**Figure. 31**). Their work is significant to researchers who are devoted to the wave or sound isolated function of auxetic foams. Such lightweight functional material is also promising in civil engineering and disaster prevention and mitigation.



621Extension (mm)622Figure. 29. Force versus central deflection for a) 3-point and b) 4-point bending test on sandwich623panels with different foam cores. Green, auxetic; blue, conventional thin; red, conventional thick. The624slope of dashed lines represents tangent stiffness of the proposed foam cores (Data replotted from625[111]).





628

Figure. 30. Cracking characteristics of (a) cementitious polymer mortar - auxetic foam composites and (b) cementitious polymer mortar-PPR fibreglass mesh composites (Adapted from [156]).



629 630

Figure. 31. The novel seismic metamaterial based on auxetic foam and the effect of Poisson's
ratio of auxetic foam on the first complete bandgap (Adapted from [157]).

For investigating and utilizing the behavior of auxetic foams under torsion, semi-auxetic rods 632 633 have been designed by Lim [158]. This novel design of rods produces unique mechanical behaviors 634 under torsion for potential use as smart structures with different responses under multivariate loading. Concentric auxetic-conventional foam rods with high modulus adhesive at the interface 635 636 have been modeled using an analytical approach to investigate the effect of the adhesive on the overall auxeticity of the system [159]. As for bending condition, where auxetic materials present the 637 characteristics of synclastic curvature, Mohanraj et al. [150] designed a hybrid composite support 638 639 for people with multiple sclerosis, consisting of an auxetic open cell foam liner and curved 640 thermoplastic plates with rhomboidal perforations. Different types of material substrates allowed in 641 this design can contribute to reducing the capital costs of development and increase the life cycle of 642 the products for these particular biomedical applications. Synclastic curvature is an ergonomic 643 deformation mode to perfectly stick a human's head, dorsum and joints (Figure. 15). In that, the 644 more uniform stress distribution provided by shape fitting could be also used as soft tissue implants 645 in cartilage articulations and meniscus repair for knee prosthetics [123].

646 Honeycomb membranes with conventional and re-entrant cell geometries have been 647 successfully fabricated and tested by Alderson et al. [160], re-entrant samples show an enhanced 648 functional performance compared to conventional (hexagonal ones) ones for glass chromatography 649 beads in applications related to filtration systems. Inspired by this unique property of auxetic 650 structures, a series of studies on auxetic foams were performed by the group of Alderson [82, 119]. 651 A variation of the pressure-drop and transport properties was also observed in auxetic foams, making 652 the negative Poisson's ratio porous material appealing for sieving of beads of uniform diameter and air pressure-drop applications (Figure. 18). The dramatic change of pore size that auxetic open cell 653 654 foams exhibit under mechanical loading (compact re-entrant pore to expanded hexagonal pore under stretching) can be useful to compensate the increase in pressure-drop which arises due to increasedfilter fouling, flushing or altering the response of a filter or catalyst structure.

The large displacements given by the auxetic behaviour for a given applied load have been also 657 highlighted by Scarpa et al. [113], and a proposal for using auxetic foams to enhance self-sensing 658 659 structural components was there presented. This large deformation feature of auxetic foams was 660 used to develop variable stiffness and shape hybrid negative Poisson's ratio PU-PE foams saturated 661 with magnetorheological fluids (MRFs). These smart foam systems have shown tunability of the 662 stiffness and magnetostrictive effects (up to 4 times stiffness increase by applying 0.2 T of magnetic 663 field) [161, 162] and tunability of the electromagnetic and acoustic absorption of surface-coated MRF foams [115]. Auxetic open cell PU-PE foams seeded with carbonyl particles and 664 665 magnetorheological fluids have also shown an increase of the acoustic absorption by a factor of  $\sim 7$ 666 compared to the pristine foam within the 1 kHz - 2.5 kHz bandwidth and the presence of a peak absorption of  $\sim 1$  with the MRF that could be shifted by almost 500 Hz when applying magnetic 667 668 fields between 0.1 T and 0.2 T [114].

As a soft material with significant lateral deformation under longitudinal loadings, auxetic foams are great candidates to serve as sensitive movement sensors. In 2013, Alderson *et al.* [163] introduced the term "piezomorphic" to describe mechanically-triggered shape change materials and proposed an elastic-gradient piezomorphic material (**Figure. 32**) made from PU foam and micro porous polymer (ex-PTFE). The piezomorphic material possesses both negative and positive Poisson's ratio regions and show a dramatic shape change triggered by the application of global and local stresses.







**Figure. 32**. Global length-width curves of different parts related to the elastic-gradient piezomorphic material (Adapted from [164]).

The piezomorphic behaviour could also be exploited to use auxetic foams as platforms to build sensors. The first contact-mode triboelectric self-powered strain sensor using an auxetic PU foam (**Figure. 33**) has been fabricated by Zhang *et al.* [164]. This sensor can be used in human body monitoring systems, self-powered scales to measure weight or in seat belts to measure the body movements on a car seat.



684Strain (%)685Figure. 33. (a) Schematics and (b) voltages under different tensile strains of the contact-mode686triboelectric self-powered strain sensor (Adapted from [164]).

687 The porous structure in auxetic foam provides the space for conductive components to 688 composite in, making auxetic foam itself with high electrical conductivity. Ahmed et al. [165] have 689 developed an AgNW (silver nanowire) - based auxetic foam sensor (Figure. 34). This device has 690 been trialed for air pressure detection and three-dimensional sensing. The AgNW-based auxetic 691 foam sensor possesses both improved piezoresistive sensitivity and stability in air or water. The 692 device also shows repeatable and reliable sensing performance under cyclic loading or unloading 693 situations. Other potential applications of this device as mentioned in the paper include sportswear, 694 safety gears, filtration and flow detection, smart healthcare foams and prosthetic liners.



695
696 Figure. 34. Schematics of (a) fabrication of the AgNW sensor and applications to (b) air pressure
697 detection and (c) three-dimensional sensing (Adapted from [165]).

698

Another interesting design of a strain-gauge sensor has been proposed on a combination of

- 699 multiwalled carbon nanotube/polydimethylsiloxane (MWCNT/PDMS) substrate fabricated on an
- open-cell PU auxetic foam by Malfa *et al.* [166] (Figure. 35). This porous sensor could be used to
- 701 integrate flexible electrode and soft robotics designs.





704

from [166]).

#### 705 6. Challenges and perspectives

706 Auxetic foams could be a potential alternative to conventional foams in applications where 707 special deformation mechanisms of the foam material play a significant role. However, most of the potential applications are limited by the substantial lack of scale-up manufacturing methodologies 708 to produce large auxetic foam samples. The development of feasible, effective and low-cost ways 709 710 to manufacture large specimens of negative Poisson's ratio foam samples is crucial to future 711 commercialization. Methodologies of utilizing chemical reactions to soften foam ribs [50, 51] or 712 decrease the required temperature during conversion [36, 53-55] are beneficial in eliminating heat 713 transfer problems. Therefore, further investigation of the chemical properties of foam materials and 714 the development of chemical conversion equipment are necessary for manufacturing scale-up of 715 auxetic foams. Some auxetic foam specimens reported in open literature do not maintain stable 716 mechanical properties in the long term, with samples also recovering their original shape after a 717 period of time, or being re-subjected to heating (shape memory effect [43, 44, 121]). One could 718 however take advantage of this feature for some specific applications (for example: if maximum 719 volume application is an issue, one could use auxetic foams in situations where the volume available 720 is greatly reduced, and make use of the shape memory effect to restore the porous material to its 721 pristine side when volume available is not anymore an issue). The long-term stability of the mechanical properties is however paramount in most engineering applications (cushions and pads, 722 723 for example). The further development of manufacturing procedures and methodologies to control 724 the recovery and stability of auxetic foams would significantly improve the use of negative 725 Poisson's ratio foams in the design at higher technology readiness levels.

726 A considerable number of geometric models with different deformation mechanisms [15, 30, 727 42, 49, 77, 79-83, 85, 87-90, 126, 134] have been proposed to simulate auxetic foams in the past 728 decades. Those models provide explanations and predictions of specific mechanical properties to 729 some extent. However, as a material with high heterogeneity [104] and anisotropy [126], auxetic 730 foams cannot be accurately described by deterministic geometric modeling only. Image-based 731 techniques like SEM and CT are able to show real and detailed micro-structure of auxetic foams 732 [36, 39, 49, 56, 73, 85, 93, 95, 96, 98]. The more systematic use of solid models extracted from  $\mu$ -CT scans and converted into FE would increase the fidelity of the modelling of auxetic foams. There 733 734 are however also some issues associated to high-fidelity FE simulations of auxetic foams extracted 735 from CT scans. The first is the need of large numbers of µ-CT volumes to be extracted from different 736 positions of the auxetic foam block. Little is known about the effective heterogeneity distributed 737 within a negative Poisson's ratio foam, scans tend to be expensive and take long time to be 738 performed, most of the data available are related to a single or very few scans inside the foam blocks. 739 The limited number of scanned volumes inside the same block of auxetic foams limits the statistical validity of the topological information that can be extracted from imaging the microstructure of the 740 741 foam itself. The second issue related to the use of FE with solid models extracted from foam scans

is the fact that very little is known about the mechanical properties of the core polymer constituting 742 743 the foams. Estimates of the Young's modulus from inverse identification of the PU in auxetic PU 744 foams subjected to mechanical loading and other data from open literature vary between 10 MPa to ~250 MPa [34, 167-169]. Moreover, the simulated stiffness and/or stress-strain curves from the µ-745 746 CT scan FE models are heavily dependent upon the type of boundary conditions and loading applied, 747 making any direct comparison with experimental curves especially obtained from homogenized auxetic foam samples quite problematic. Any technique developed to increase the number of high-748 749 fidelity scans in an affordable way and improve the reliability of the FE modelling in terms of core 750 material properties and boundary conditions would significantly help the computational modelling 751 of auxetic foams. Another approach that could offer some advantages in modelling statistically 752 realistic auxetic foams is the use of 2D random models that show some similarity to the topology of 753 negative Poisson's ratio cellular materials like Thiessen polygons (Voronoi diagrams) [170-173]. 754 Quite interestingly, Li et al. [91] have shown that by using 2D Voronoi networks to simulate Cu 755 copper foams subjected to strain hardening and biaxial compression, it is possible to predict the 756 resulting Poisson's ratio and EA capacity of the real auxetic metal foams. The use of stochastic 757 based tessellation for PU pristine foams based on Kelvin lattices or similar topologies and more in-758 depth information about the mechanics and shape memory effect of the core PU could be 759 instrumental to create a digital twin of the manufacturing process of auxetic foams. Lakes et al. also 760 introduced Cosserat (micropolar) theory [174] for the analysis of materials with negative Poisson's 761 ratios [108, 175, 176], stating that syntactic foams could be regarded as classical ones apart from 762 small deviations. Rueger and Lakes however found compelling experimental evidence of the 763 existence of Cosserat torsion lengths and coupling due to size effects while testing the foams [177]. 764 Gaspar et al. [178] developed a theoretical framework to predict the elastic properties of a material 765 with contacting microstructure originally proposed by Koenders [179] and extending the previous 766 formulation to continuum materials with auxetic behaviour and anisotropic heterogeneity. Ciambella et al. [180] and Rueger et al. [181] modeled auxetic open cell foams as continuum solids 767 768 for the analysis of nonlinear elasticity within a modified Ogden hyperelastic anisotropic framework. 769 Recently, the group of Montáns [182, 183] provided a series of extension of phenomenological linear theory, realizing the simulation of strain-stress relation and auxetic behaviour of auxetic foam 770 771 through an accurate numerical methodology. Such methodology makes orthotropic materials not 772 only feasible but also efficient (compared with FE analysis) simulated. However, the simulation processes of their works are carried on axial quasi-static loading only, some sophisticated loading 773 774 conditions such as impact, crushing and cyclic loading are inescapable in applications, therefore are 775 desirable to be simulated as well. Auxetic foams produced with the current manufacturing 776 methodologies are difficult to simulate with high-fidelity continuum Cauchy classical elasticity 777 models. The theoretical approaches discussed above are an indicator for the need to further develop 778 more high-fidelity continuum theories and therefore to realistically simulate the deformation 779 mechanisms and size effects observed in real foams.

780 Another interesting and promising concept is to engineer the large lateral deformations triggered by axial loading in piezomorphic foams to design and fabricate sensors [163-166]. Soft 781 782 sensors have significant potential in applications like soft robotic, biomedical and behavioral 783 sensing, smart belts (Figure. 36) and smart clothing. Piezomorphic materials [34] could be also used 784 as actuator elements by enabling turning on/off of devices in response to stress-induced shape 785 changes (Figure. 36). Other potential applications are in the smart bandage sector (drugs can be released when the bandage is stretched by the swelling of the wound), adaptive airfoils (stress 786 induced shape change), prosthetic limb lining (fit stump volume variations), bra cups (expand during 787 788 vigorous exercise and/or periods of significant variations in breast size) and deployable/removable 789 cores (easy to insert and hard to extract, a similar application of auxetic nails can be seen in ref. 790 [184]).





793

**Figure. 36**. Examples of potential applications utilizing the unique stress-induced shape change of auxetic foam materials (middle): smart belts (upper) and actuator elements (lower).

794 Composite materials and structures with auxetic foams are also promising for applications like 795 complaint composites for prosthesis, impact and blast protection. Recently, composite structures 796 with auxetic foams and cellular materials have been designed, modelled and built, showing 797 interesting performances for energy [141, 152, 185] and shear resistance [19, 31, 38, 45, 83, 108, 798 109, 111]. Compared with other traditional core materials, auxetic foams tend however to possess a 799 low stiffness. It would be therefore interesting to develop more rigid and lightweight auxetic foams 800 with enhanced mechanical properties, both on the polymeric and metal foam side, as well as to 801 explore hybrid combinations of auxetic foams and other more rigid materials or structures. 802 Lightweight characteristics are also important. Auxetic foams are denser than their pristine conventional counterpart used for the production, depending on the volumetric compression ratio. 803 While some specific mechanical properties are significantly enhanced compared to the conventional 804 805 original foam, the comparison of the specific properties (i.e., density or weight averaged) between 806 auxetic and conventional foams may not offer the same advantages as the direct ones. Besides, as a 807 type of cellular material, auxetic foam is potential to be a candidate for matrix material, in which fluid or powder additions, such as graphene oxide [117] and Er [121], of which are easy to disperse 808 809 and adhere to the foam ribs, i.e., forming composite foam materials (Figure. 37). Composite foams 810 hold the superior properties provided by additional materials meanwhile retain the properties of 811 foam materials themselves, many composite foams are still based on conventional foams [186-194], 812 it is potential to convert those composite foams into auxetics, in that original foams could obtain 813 triple functions.







A series of configurations made using auxetic frames and inclusions of conventional foam have been also investigated (**Figure. 38**) [195, 196]. In a broad sense, these configurations can be defined as hybrid auxetic by increasing the bulk properties of the composite with the foam matrix and using the auxetic lattice in a synergistic way to increase the EA and impact protection.





In addition, rigid open-cell auxetic and metal foams could be used as scaffolds to build silicone [197], mortar [156], concrete and other similar high-performance composites. Random reticular structures of the foam scaffolds could contribute to oppose shear failure and fracture in these foambased composites [156].

Another aspect to be considered and of increasingly significant importance is the environmental impact of auxetic foams, especially in view of scale-up manufacturing processes. All the manufacturing processes used to fabricate polymeric foams, in particular, necessity of either temperature and/or use of chemicals. Considering the contribution of tooling, molding and labor, detailed life cycle costs of the different manufacturing processes are needed to make the business and environmental case for the large-scale manufacturing of auxetic foams. The use of more biobased polymers and products during the manufacturing is also an interesting aspect to be considered.

#### 836 **7. Conclusions**

In this paper, the state-of-the-art of manufacturing, characteristics and applications of auxetic 837 838 foams have been reviewed. Existing methodologies for the conversion of auxetic foams have been 839 presented and focus has been put on the relations between the different possible manufacturing 840 parameters and recommended optimal combinations of those. The micro-structure and deformation 841 mechanisms in auxetic foams have been discussed, from theoretical unit cell models to more high-842 fidelity numerical ones based on geometry information extracted from imaging techniques. The 843 mechanical properties of auxetic foams have been considered here in terms of stiffness, hardness 844 and toughness, showing superior performance in loadings like indentation, impact, cyclic and 845 vibroacoustic. Auxetic foams could represent enhanced alternatives to conventional foams in many 846 applications where unusual deformation mechanisms of the foam material play a role. We have also 847 provided a consistent number of examples of novel applications of auxetic ranging from biomedical, aerospace, filtration and sensing, to name a few. This paper also provides a discussion of existing 848

- challenges and future research directions to enhance the feasibility and the design space of auxetic
- 850 foam materials and structures.

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