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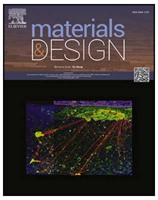

3D printing of active mechanical metamaterials: A critical review

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ABSTRACT

The emergence of mechanical metamaterials from 4D printing has paved the way for developing advanced hierarchical structures with superior multifunctionalities. In particular, 4D-printed mechanical metamaterials exhibit extraordinary mechanical performance by integrating multiphysics stimuli with advanced structures when actuated by external factors, thereby altering their shapes, properties, and functionalities. This critical review offers readers a comprehensive overview of the rapidly growing 4D printing technology for developing novel mechanical metamaterials. It provides essential information about the multifunctionalities of 4D-printed mechanical metamaterials, including energy absorption and shape-morphing behavior in response to physical, chemical, or mechanical stimuli. These capabilities are key to developing smart and intelligent structures for multifunctional applications such as biomedical, photonics, acoustics, energy storage, and thermal insulation. The primary focus of this review is to describe the structural and functional applications of mechanical metamaterials developed through 4D printing. This technology leverages the shape-shifting functions of smart materials in applications such as micro-grippers, soft robots, biomedical devices, and self-deployable structures. Additionally, the review addresses current progress and challenges in the field of 4D-printed mechanical metamaterials. In conclusion, recent developments in 4D-printed mechanical metamaterials could establish a new paradigm for applications in engineering and science.

1. Introduction

Recently, 3D printing has completely revolutionized modern manufacturing by redefining how objects are conceptualized, designed, and created [1]. Moreover, its rapid development over the past decade has triggered the development of functional components for virtually all manufacturing sectors due to its design flexibility and rapid prototyping capabilities [2,3]. 3D printing produces complex 3D structures by adding material in small volume elements called voxels [4]. In the meantime, this technology has found diverse and novel applications in aerospace, construction, electronics, and biomedical sectors, thanks to the development of advanced multifunctional materials [5]. Commonly used additive manufacturing (AM) techniques for developing complex designs through layer-by-layer strategy are namely fused deposition modelling (FDM) [6], stereolithography (SLA) [7], digital light processing (DLP) [8], selective laser melting/sintering (SLM/SLS) [9], direct ink writing (DIW) [10], electron beam melting (EBM) [11], inkjet printing (IJP) [12], and material jetting (MJ) [13]. Over time, these printing approaches are becoming more accessible and cheaper. Each 3D printing technique has its benefits and drawbacks in terms of precision dimensioning, reproducibility, building time, surface finish uniformity, mechanical properties, and material cost [14]. However, the primary drawback of 3D printing technology is its inability to produce shape-morphing and adaptive products [15].

Metamaterials, which are usually termed artificially engineered materials, are relatively a new class of well-known materials for their ability to achieve complex properties [16]. Specifically, metamaterials with periodically arranged patterns have garnered wide praise from the scientific community due to their novel and exceptional features [17–19]. Their low weight and remarkable structural and functional properties have set the stage for cutting-edge applications [20], which are detailed in subsequent sections. Reconfigurable metamaterials are

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comprised of tunable elements or deformable structures, which can transform into various shapes under any environmental changes such structures require the shape and size of the building block in the design domain eliminating any homogeneous requirements [21]. Fig. 1 summarizes typical keywords associated with metamaterials. Using traditional manufacturing processes, it is very challenging to develop fully functional metamaterials because their internal structures require increasingly complex designs at the micro-scale [22]. Moreover, their configurations and properties cannot be changed through these traditional manufacturing methods, which limits their further development and adaptation to the particular geometric needs of various applications [23].

3D printing is well known for creating complex parts with high precision, allowing the integrated fabrication of complex and multiscale (macro and micro) metamaterials [24]. This technology has advanced the development of metamaterials, surpassing traditional materials in many engineering applications [25]. Also, It has inspired researchers to impart additional multifunctionalities such as electromagnetic manipulation, programmable morphability, shape memory behavior, topological and electromagnetic absorbers in aerospace engineering [26–28]. Today's 3D-printed mechanical metamaterials exhibit impressive properties such as high strength density ratio, negative compressibility, negative or zero Poisson's ratio (auxetics), dynamic stiffness, stiffness under volumetric compression but softness under shear load, and frequency-dependent mechanical properties. Moreover, these metamaterials are enriched with compact size and small thicknesses with shape-shifting abilities [29], making them advantageous in biomedical, space and automotive applications [30,31]. Recently, metamaterials have witnessed unprecedented advancement regarding their geometrical, process and functional innovation leading to their widespread applications with complex and tailored geometries and improved multifunctional properties. Fig. 2 provides an overview of all these innovations including functional innovations (Fig. 2(a–c)), process innovations mainly due to 3D printing (referring to Fig. 2(d–f)) and geometrical features (Fig. 2(g–j)) for many intriguing applications. Nowadays 3D printing and various artificial intelligence (AI) tools work collaboratively to explore novel codes and optimization tools for decoding complex correlations for realizing more intricate and realistic structures, while continuously improving the design structure by comparing from the well-established AI dataset [32].

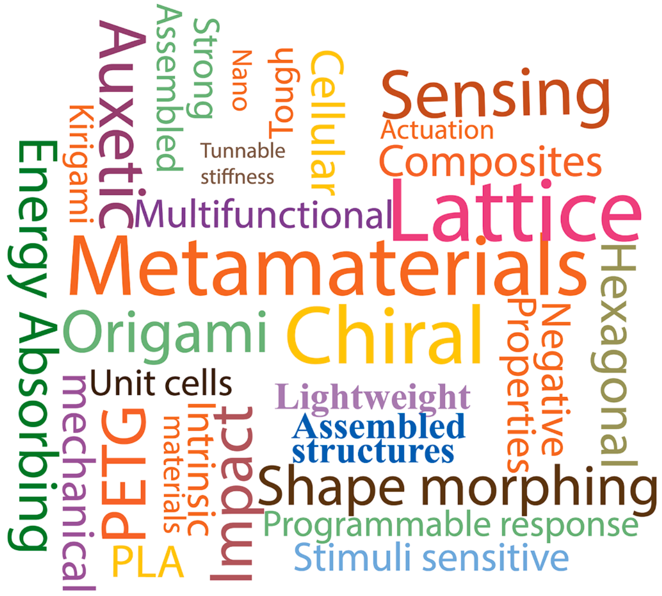

Fig. 1. Summary of keywords associated with metamaterials (these keywords appear in the title or as separate keywords while searching for 3D/4D printing of mechanical metamaterials during this current review planning and organization from the Scopus database).

Thanks to intense efforts by research communities across various disciplines, the emergence of intelligent materials and 3D printing technology now allows for the creation of functional and adaptive structures that traditional 3D printing cannot achieve [43]. This cutting-edge material printing approach, known as four-dimensional (4D) printing, incorporates a 4th dimension to traditional AM technology, inducing shape-morphing behavior in smart materials and uncovering new horizons in manufacturing intelligent systems and smart structures [44]. Smart materials possess many exciting capabilities, including shape memory, self-sensing, self-actuating, and self-assembly [45]. To date, the prime focus of 4D printing technology has been on shape-memory materials [46]. These materials possess the ability to produce a shape-morphing effect upon exposure to certain stimuli and return to their original position after the stimuli are removed [47]. Broadly speaking, 4D printing generates changes in shapes, properties, and functionalities that can evolve over time in the presence of one or more stimuli [48]. The technology uses a wide range of intelligent materials such as shape memory polymers (SMPs), shape memory alloys (SMAs), shape memory ceramics (SMCs), various gels, liquid crystal elastomers (LCEs), and other soft hybrid materials that respond to external stimuli like temperature, electric field, light, chemical solvent, moisture, magnetic field, etc. [49]. Consequently, 4D printing directly fabricates interactive and intelligent biomedical devices, aerospace components, electronics parts, metamaterials, controllable structures, smart actuators, soft robots, and other multifunctional devices. Additionally, it offers certain benefits such as developing robust, flexible, and deformable smart architectures, increased manufacturing efficiency, and cost reduction [50].

According to Kuang et al. [51], 4D printing is one of the top innovative and emerging technologies in the world and is expected to reach its zenith in the next few years. Additionally, a revolutionary strategy combining metamaterials with 4D printing is advancing towards creating ground breaking artificial structural materials by breaking current limitations. Metamaterials are well-known man-made structures that are now continuously evolving to fulfil human needs. We believe this review consolidates the latest progress in 4D-printed mechanical metamaterials and provides valuable insight into this exciting field. The primary focus of this review is to shed new light on the multi-functionality of 4D-printed mechanical metamaterials as summarized in Fig. 3. This will be followed by a discussion of critical factors, such as potential smart materials, printing techniques, and emerging applications, with closing remarks on how to integrate artificial intelligence approaches to develop more practical designs for these structures. In this regard, many researchers have attempted to summarize the essential knowledge about metamaterials. For instance, Kowerdziej et al. [52] review active metamaterials utilizing resonances in plasmonic and dielectric materials hybridized with soft-matter assemblies such as colloids, liquid crystals, granular matter and polymers. Sinha and Mukhopadhyay [53] provided an in-depth review of artificially engineered mechanical metamaterials, with a primary focus on their active, reconfigurable and multi-physical behavior only. Zhou et al. [54] discussed 3D/4D printed mechanical metamaterials in terms of manufacturing and applications, their review primarily focuses on applications. Lastly, Isaac and Duddeck [55] explored the multifunctional features of all metamaterials mainly from smart materials and geometric point of view only. However, this current review highlights recent advances specifically in 4D-printed mechanical metamaterial technologies, emphasizing their multifunctionalities, including shape-morphing abilities, and energy-absorbing characteristics. Additionally, we spotlight the latest research on design optimization using novel AI tools from the last two years. We recommend new horizons for 4D-printed mechanical metamaterials by highlighting advanced printing techniques and smart materials to fully exploit their potential in the 4D printing realm. We adopted a systematic approach. First, we discussed briefly 4D printing technology in Section 2. Next, Section 3 includes an overview of 4D-

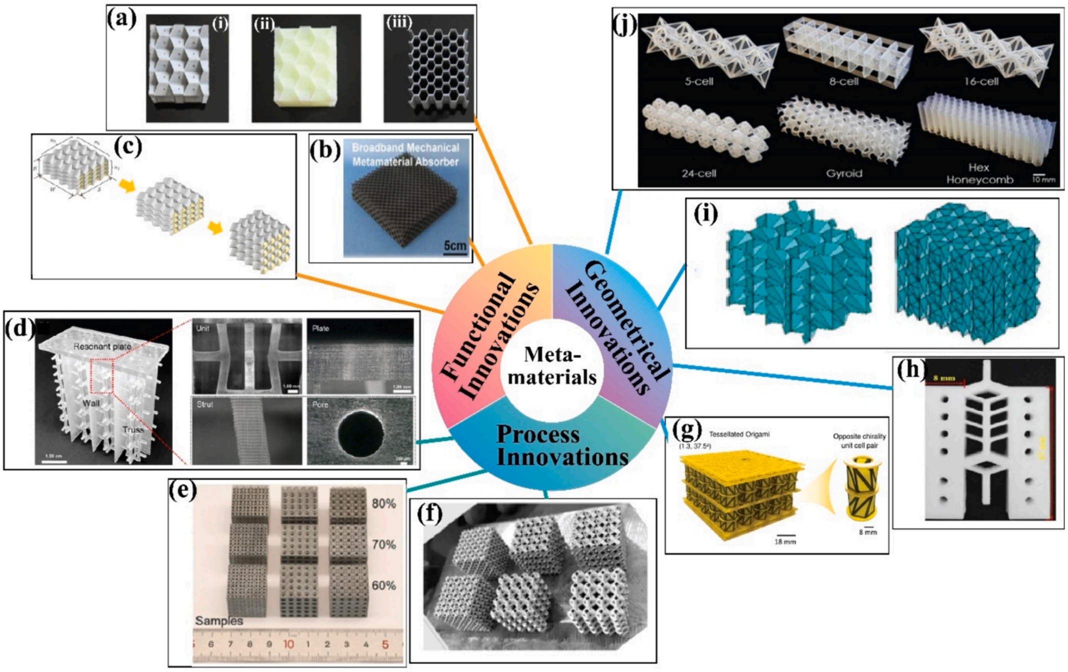

Fig. 2. Metamaterials innovations: (a) Buckling-regulated origami materials ((i) 316L stainless steel, (ii) PC plastic and (iii) hexagonal honeycomb made of 316L stainless steel) with synergy of deployable and undeployable features (adapted from Ref. [33] copyright 2023 Elsevier Ltd.); (b) 3D-printed broadband mechanical metamaterial absorber endowed with dual-functionality of electromagnetic wave absorption and reinforced relative stiffness (adapted from Ref. [34] copyright 2022 Elsevier B.V.); (c) Continues shape morphing mode of the curved crease origami metamaterial comprising of n_1 stacked unit cells in the in-plane transverse direction, n_2 in the in-plane longitudinal direction and n_3 in the stacking thickness direction (adapted from Ref. [35] copyright 2023 Elsevier Ltd.); (d) Decoupling-enabled porous multifunctional metamaterials with microstructural characteristics: the re-entrant unit, resonant plate, strut, and micro-perforation (adapted from Ref. [36] under Creative Commons Attribution-Non Commercial 3.0 Licence); (e) Voronoi-based body-centered cubic, Voronoi-based regular octahedral cubic, Voronoi-based body- and face-centered cubic-based metamaterials fabricated via LBF 3D printing process for bone implant applications (adapted from Ref. [37] copyright 2024 Elsevier Ltd.); (f) Novel three face-centered cubic (FCC) lattice-based mechanical metamaterials, inspired by atomic packing and the hollow features of bamboo, developed through SLM of Ti-6Al-4V with high fidelity (adapted from Ref. [38] copyright 2021 Elsevier Ltd.); (g) 3D-printed tessellated origami-based material with a pair of opposite chirality unit cells (adapted from Ref. [39]); (h) Novel mechanical metamaterial based on a fishbone-like structure with polar and dual deformation characteristics, allowing surface structure to be hard while the opposite side remains soft, allowing task adaptation to different load levels on the soft side (adapted from Ref. [40] copyright 2023 Elsevier Ltd.); (i) The geometrical configurations of two types (wall replaced and wall added) of origami-embedded honeycombs structures for improved energy absorption performance (adapted from Ref. [41] copyright, 2023 Elsevier Ltd.); (j) Cubically symmetric mechanical metamaterials from three-space geometrical shadows of 4D geometries (4-polytopes) with various cells (figures are arranged from left to right) such as 5-cell, 8-cell, 16-cell, and 24-cell, and extra structures such as gyroid, and hexagonal honeycomb employed as "comparative experimental controls" (adapted from Ref. [42] copyright 2023 the Authors published by American Chemical Society).

printed mechanical metamaterials with special consideration on design optimization through novel AI tools in Section 4. Section 5 details the main focus of the review, specifically the shape-morphing and energy-absorbing abilities of 4D-printed mechanical metamaterials. Booming applications of 4D-printed mechanical metamaterials as structural materials, smart actuators, novel biomedical devices and self-deployable structures are outlined in Section 6. Finally, the perspective on advances in each 4D printing process, their stimulation mechanism, shape-morphing abilities, and the integration of multifunctionalities in mechanical metamaterials are proposed in Section 7.

2. 4D printing

4D printing uses AM technology, which permits the printing of intelligent materials directly through a computer-aided design (CAD) to develop smart and intricate products with sizes ranging from micrometres (even nanoscales) to meters [56]. Notable 3D printing techniques such as FDM, SLS, SLA, SLM, DIW, and EBM are employed for the 4D printing of materials, depending upon the compatibility with the 3D printer [57]. DIW and FDM are extrusion-based techniques, which extrude ink from a nozzle and print 3D objects by moving in a specified path. Multi-material printing can also be performed by using extrusion-based printers with two or more printheads, which deposit different inks during a single print job [58]. A variety of thermoplastic materials used in the FDM technique are unsuitable for 4D printing due to their incompatibility with intelligent materials [59]. SLA, DLP and two-photon polymerization (TPP) fall into the category of vat-photopolymerization, in which light of specified wave lengths is used to polymerize various monomers. Out of these techniques, TPP permits the fabrication of extremely intricate and precise objects with high spatial resolution [60].

The 4D printing technology requires intelligent materials, AM technology, and suitable stimulus [61]. Therefore, it is essential to consider appropriate stimuli and materials to develop smart structures through the 4D printing technology [62]. A variety of stimuli including chemical (pH, moisture, or redox state of metal ions), physical (light, temperature, electric field, or magnetic field), biological (cell traction force, biomolecules, or enzymes), or their combination are used simultaneously or sequentially to induce permanent or temporary shape change in smart or programmable materials [63]. Some intelligent materials also exhibit color-changing properties in response to certain stimuli such as light [64]. Self-healing ability is another important feature, which has become pivotal in 4D printing technology. This property allows smart materials to enable function recovery and structural restoration upon damage, which enhances the material's reliability [65].

Smart materials undergo different mechanisms such as constrained or un-constrained, upon exposure to the external environment. A constrained mechanism occurs when materials cannot retain their original states after deformation. On the contrary, an un-constrained mechanism

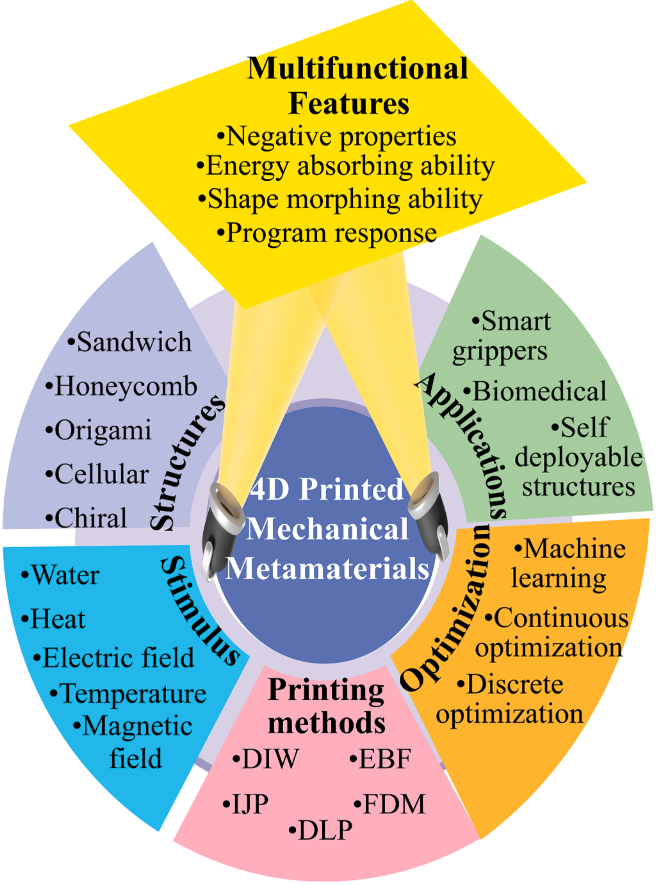

Fig. 3. An overview of 4D-printed metamaterials, with particular emphasis on their multifunctionalities covered in the current review theme.

helps materials regain their original shapes [66]. These mechanisms trigger shape-changing, shape-memory, and self-healing effects [67]. The shape-changing phenomenon under the response of some external stimuli is one of the major characteristics of the 4D printing which differentiates this technique from the traditional 3D printing processes [68]. Smart materials utilized in 4D printing change their shape according to the given environmental stimuli that trigger the macromolecular movements, in the form of shrinkage, stretching, swelling, rolling, folding, bending, and twisting [69]. The shape-memory or shape-morphing effects that existed in 4D-printed objects comply with external stimuli. Shape-morphing effect produces irreversible changes in the properties of objects, whereas, the SME in programmable materials results in the transition between the temporary and permanent shape(s) when exposed to an external stimulus [70]. Based on morphological transformations, SME of programmable materials is further divided into irreversible one-way SME and reversible SME. Besides low-end applications, these SMEs are also highly beneficial in developing self-deployable hinges and smart biomedical devices [71]. As reported by Zhang et al. [72], SME is not an intrinsic characteristic of SMP materials; it is developed through thermomechanical training procedures as well as molecular architecture (see Fig. 4). Four stages are generally used for illustrating a shape memory cycle of thermal responsive. The first stage is programming, followed by fixing, then unloading and finally, recovery. When the segments are reheated to Trec (above the phase transition temperature), the stored energy is released, driving the net-points to return to their memorized positions. Most 4D-printed shape memory structures are reheated and recovered freely to their original shape without any constraint, a process known as free recovery.

3. Mechanical metamaterials

Metamaterials are defined as artificial materials with engineered architectures and have emerged as a novel research frontier in chemistry, physics, engineering, and material science [73]. These materials offer progressive prospects in mechanics, thermotics, acoustics, electromagnetics and optics [74]. One of the most significant features of metamaterials lies in the engineered structures of their unit cells, rather than the intrinsic properties of the materials used [75]. They are designed based on the geometry of the “cellular” structure, irrespective of the chemical composition, allowing them to achieve extraordinary physical properties (acoustic, optical, thermal, mechanical, electronic, etc.) that are often lacking in natural or chemically synthesized

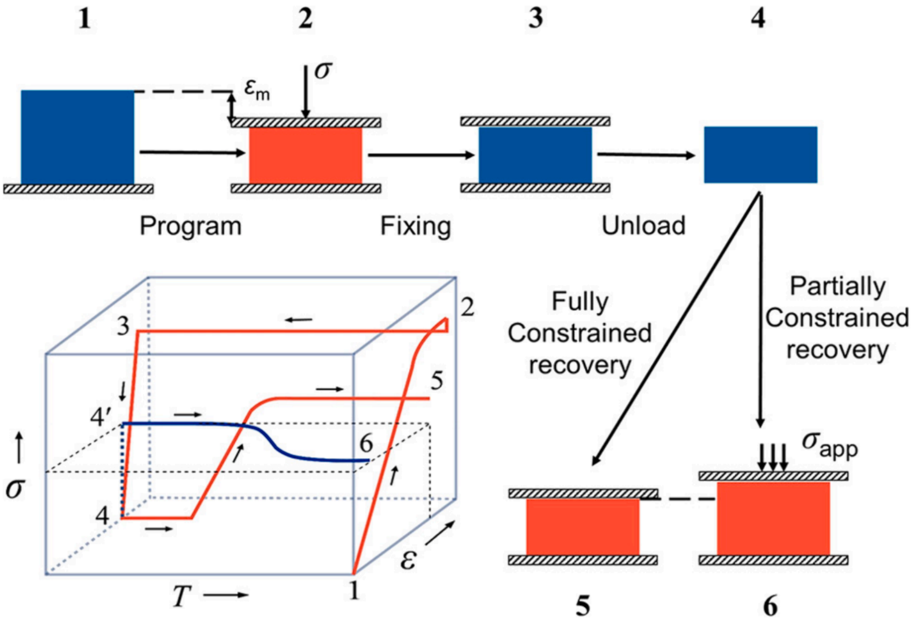

Fig. 4. Demonstration of constrained shape recovery and the relationship among stress (σ), strain (ε) and temperature (T) during thermomechanical testing. The process involves loading at high temperature (curve 1–2), followed by fixing (curve 3–4) and unloading at low temperature (curve 2–3). Fully constrained shape recovery is represented by curve 4–5, while partially constrained shape recovery is indicated by curve 4–6 [72].

materials [76]. In recent decades, researchers have developed novel 3D-printed mechanical metamaterials, by designing accurate, multimaterial microstructures with low density, and high stiffness. This has opened new avenues for building metastructures with various auxetic properties, including negative stiffness, negative Poisson's ratio (NPR), and quasi-zero stiffness multifunctional characteristics [77]. The macroscopic responses of 3D-printed mechanical metamaterials under applied forces can be programmed by unit cell design to control various characteristics such as shape matching, NPR, and shape morphing [78]. Novel intelligent design, such as origami/kirigami (cutting/folding papers into complex 3D or 2D objects) and bistable structures, makes these metamaterials more practical, innovative, and reconfigurable. Most of these advancements come from 4D printing, which holds significant practical importance [79]. Particularly, advanced functionalities have been integrated into their texture through the interaction between material and structure, positioning them as mechanical metamaterials that serve as building blocks for future intelligent matter. This adaptability extends to other fields as well, such as energy harvesting, actuation, and adaptation, to name just a few [80].

There are various fundamental mechanical characteristics associated with basic mechanical metamaterials such as auxetic, lattice, pentamode, chiral, origami, and kirigami structures [81]. 3D-printed auxetic metamaterials, such as perforated structures, have captured significant attention in automotive engineering, sports protection, and aerospace for their stable deformation characteristics and excellent energy absorption capabilities [82]. In lattice-based metamaterials, periodic unit cells are arranged in lattices to achieve unique mechanical properties, such as an NPR in re-entrant auxetic structures and an ultra-high strength-to-weight ratio in hexagonal honeycomb lattices. Emerging origami folding techniques, such as folding and unfolding, facilitate the design of origami metamaterials with multifunctional mechanical characteristics, including controlled stiffness and shape-morphing abilities. To date, patterns like Yoshimura pattern, Miura-ori pattern, and waterbomb origami have been developed. Likewise, triangle-based kagome patterns and rotating squares patterns have been studied for their mechanical and geometrical properties in designing the kirigami structures. This has been demonstrated by architected materials based on origami engineering, which have shown the enormous potential of metamaterials in medical devices, deployable structures, and programmable matter applications. These materials achieve multifunctionalities, including vibration isolation, and shape morphing [83]. Optimization approaches, discussed in the next section, facilitate the systematic and efficient development of various structures with tuned stiffness, multistability, shape morphing, and mechanical properties.

3.1. Smart materials for metamaterials

Smart materials (SMs), a class of stimuli-responsive materials (SRMs) are generally used for 4D printing thanks to their unique programmability properties and tailored deformation with specific designed stimuli patterns [84]. These programmable materials include magneto-active alloys, magneto-active polymers, light-responsive polymers, temperature-responsive polymers, humidity-responsive polymers, pH-responsive polymers, and electro-active polymers [85]. These materials exhibit self-healing, self-assembly, and shape-memory behaviors in response to external stimuli, which make these materials prominent in myriad engineering sectors. Simple integration, low material, and fabrication costs are some of the key advantages of these smart polymer composites [86]. Currently, thermally programmable liquid crystal polymers, SMPs, stress-relaxing filaments, magnetic materials and SMP composites, are most extensively applied for developing engineered metamaterials through 4D printing. Mostly, the research in 4D printing focuses on shape-morphing abilities like corrugation, bending, elongation, folding, swelling, and twisting of smart materials [87]. Moreover, synthetic low-density polymers such as polycaprolactone (PCL), polyurethane (PU), polylactide (PLA), and polyethylene terephthalate glycol capable of lowering the damage triggered by shock due to impact loading are of great interest in developing auxetic metamaterials [88,89]. Sometimes, the addition of different fillers such as iron oxide (Fe_3O_4) and carbon nanotubes (CNTs) allows the polymers during the 4D printing to help in retaining metamaterial excellent shape morphing capabilities.

3.2. 4D-printed mechanical metamaterials

The composite structures that go beyond the capabilities of their individual constituents (by adding physical controls), and whose effective properties may even opposite those of their constituents are broadly classified as stimuli-responsive metamaterials or 4D metamaterials (referring to Fig. 5) [90]. The programmable shape morphing with controlled mechanical properties of 4D-printed mechanical metamaterials can be as simple as that of an individual bit, as discussed in the literature. For instance, it can be best described as a bistable mechanism that switches between two configurations, a dynamically excited system that changes phase in response to an input signal, or a more complex integrated network [91]. 4D-printed mechanical metamaterials is a flourishing field that continuously advances through mechanisms such as buckling, rotation, or folding of these building blocks. These mechanisms achieve counterintuitive characteristics and functionalities, including shape morphing, programmability, and tunable mechanical responses. Additionally, the expanding capabilities of 4D printing enable mechanical metamaterials to surpass the inherent characteristics of their constituent materials, exhibiting remarkable performance with unique characteristics for many engineering applications.

Innovative structural designs are continuously improving material capacity not only to improve the load-bearing ability but also to better adapt to the changes in the environment and multifunctional structures for fulfilling the demand of the industry where structures often need to face harsh environmental conditions such as earthquakes by improving vibration isolation performance [93,94]. It is crucial for innovative structural design apart from lightweights must have necessary functionalities which can respond well to such environmental conditions [95]. In this regard, unconventional mechanical metamaterials, with tunable material-structure–function design are considered vital breakthroughs for designing advanced structures, with intelligent and bionic features for paving the future of mechanostructures for advanced structural application [96]. Moreover, traditional materials, when joined for structural applications, often have different coefficients of thermal expansion (CTE), resulting in structural distortion and potential failure. In contrast, mechanical metamaterials with customized CTEs are gaining attention for their ability to produce isotropic CTEs [97,98]. This isotopic CTE in mechanical metamaterials is achieved by rationally controlling cellular microarchitectures and specific material layouts, which helps to facilitate thermal mismatches within internal beams or struts [99,100]. This approach generates tunable CTEs at a macroscopic level, mitigating large thermal deformations and stresses that could otherwise lead to structural failure. 4D-printed mechanical metamaterials are periodically arranged by microstructures to show excellent mechanical properties for matching the demand for reliable structural materials. It is worth noting that at the microstructure level, the localized behavior of these materials is structure-like, and is considered almost homogenized, resulting in improved multifunctional performance [30]. Mechanical metamaterials with programmable CTEs are referred to as stretching-dominated and bending-dominated [101]. According to Yang et al. [102], 4D-printed metamaterials are extremely lightweight and demonstrate superior multifunctionalities, such as geometrical reconfigurability, functional deployability, and ultra-tunable shock absorption under high-impact loading. These metamaterials can be significantly deformed and mechanically programmed into arbitrary shapes, with the ability to fully regain their original shape at any time, as shown in Fig. 6(a). These micro-architected programmable metamaterials have promising applications in a wide range of

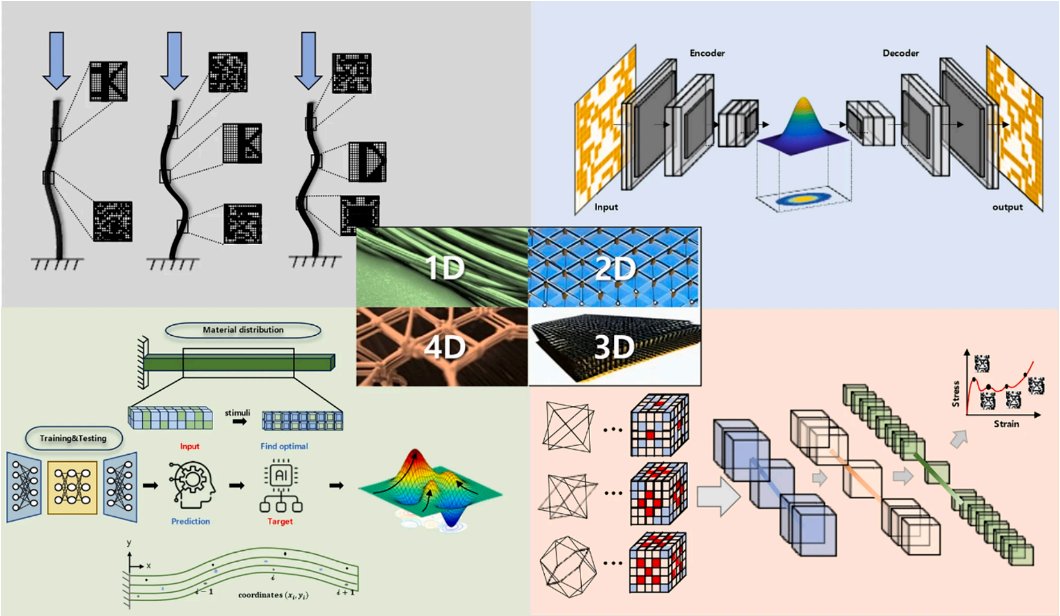

Fig. 5. An overview of metamaterial design from 1D to 4D (adapted from Ref. [92] copyright 2023, the Author(s), published by Korean Society for Precision Engineering, Springer Nature).

fields, including morphing aerospace structures, implantable biomedical microdevices, soft robotics, and tunable shock-absorbing interfaces.

Apart from the high structural applications of 4D-printed mechanical materials, recent research on 4D-printed metamaterial also focused on the programming of shape changes structures with various stimuli. These structure variations referred to as shape-morphing structures are highly attractive for intelligent machines in various interdisciplinary fields [103]. Recently, Bai et al. [104] introduced FDM-based moisture-sensitive mechanical metamaterials utilizing bi-material curved strips that exhibit repeatable programming capabilities for demonstrating customized and unusual hygroscopic deformations. The study showed that 4D-printed metamaterials exhibited unusual hygroscopic deformation modes, including anisotropic, shearing, gradient, bending, and 3D deformation of 2D structures, as illustrated in Fig. 6(b). These deformations featured a tunable coefficient of hygroscopic expansion ranging from negative to positive, depending on material configurations and the structural design of geometrical parameters. A pixelated design and coding of the building blocks enable programmable metamaterials to achieve arbitrary hygroscopic deformations. Additionally, this hygroscopic deformation can be reprogrammed by applying erasable moisture-proof coatings to specific areas of the metamaterials, allowing for the continuous customization of different deformation modes.

4D-printed mechanical metamaterials offer exciting possibilities for developing novel structures for energy storage applications by incorporating shape-changing materials to optimize their elemental configuration or internal structure [105]. This enables smart structures to improve their energy storage capabilities and, consequently, their impact performance. These advancements can contribute to the development of more efficient and compact structures for energy-efficient and portable devices in flexible electronics and self-deployable structures [106,107]. Thus, improving overall structure efficiency and resilience by adapting to both anticipated and unexpected fluctuations during their service life. Recently, Yao et al. [108] showed that 3D programmable resilient mechanical metamaterials (PRMMs) are designed to form curved beams with chiral metastructures without relying on flexible or hyperelastic constituents. These metamaterials showed nonlinear resilience and anisotropy, with large elastic compression strains (>0.75) and significant programmable modulus reduction. Additionally, a soft sample was dropped multiple times from a height of one meter, and its dynamic response was monitored using a high-speed camera to assess the reliability and elasticity of PRMMs under impact. As highlighted in Fig. 6(c), a solid sphere (56 g) dropped from a height of 1 m and reached a maximum rebound height of only 0.48 m after contacting a rigid surface. Thus, the modular compliant cube absorbed nearly all mechanical energy through elastic contraction. It was found that the impact energy was localized within a specific region of the meta-plate, suggesting its exceptional properties in energy dissipation. Likewise, Zeng et al. [109] designed 4D-printed stair-stepping mechanical metamaterials with programmable load plateaus. They presented three typical design strategies for customizing the number of load plateaus and quasi-zero stiffness (QZS) characteristics on the force–displacement curve, enabling the creation of both single and multiple QZS-zone metamaterials with adjustable plateau width and stair-stepping plateaus respectively. Additionally, two reversible methods—shape memory programming and supporting payload—were introduced for switching among various unit configurations, as presented in Fig. 6(d). This study offered a promising approach for designing programmable load plateaus and developing novel metamaterials with tunable force–displacement responses for innovative vibration isolation applications.

The remarkable mechanical and shape memory performance such as high shape recovery force and the shape recovery ratio of the 4D printed mechanical metamaterials is setting their role in functional applications. For instance, Yue et al. [110] investigated the self-expanding and self-folding performance of novel thick-walled 3D kirigami-inspired and honeycomb structures driven by 4D printing. This study demonstrated that these 4D printed structures exhibited excellent programmability and shape memory ability, as shown in Fig. 6(e). Moreover, during this shape change behvaiour, these structures showed a large volume change ratio, which can effectively recover load-bearing capacity when the angle is positive, making them ideal for space-saving smart load-bearing equipment applications. Wang et al. [111] developed a versatile fabrication-design-actuation strategy for accurately controlling electrothermal origami with remarkable mechanical properties and spatiotemporal controllability using FDM. The study provided insights into the controllable actuation performance of the proposed 4D-printed electrothermal origami, achieved through Joule heating, which enhanced actuation force due to improved heat conduction, and mechanical characteristics by incorporating continuous carbon fibers as reinforcement. Additionally, the proposed electrothermal active origami can be reconfigured and locked into any desired configuration by regulating

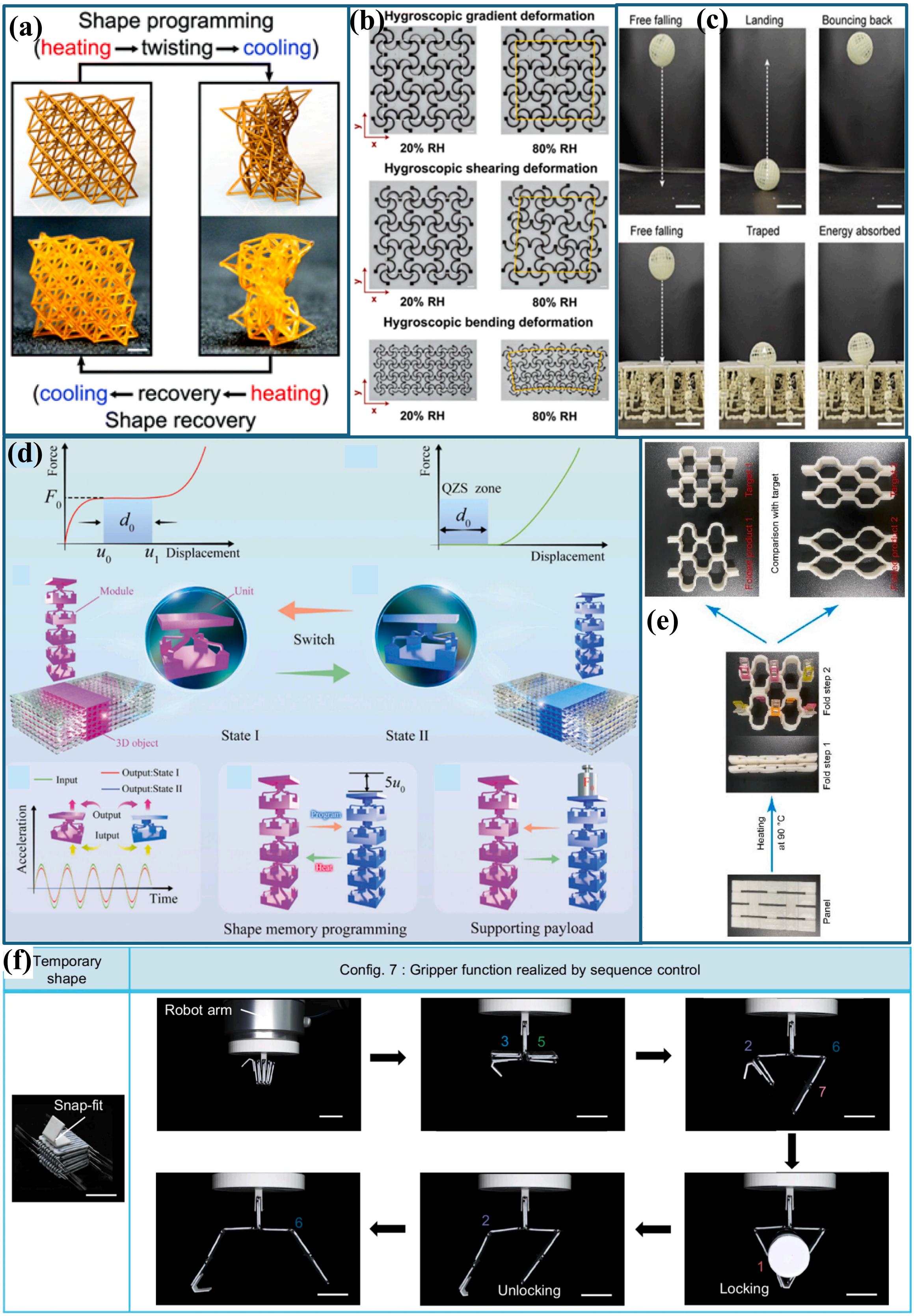

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Fig. 6. (a) Demonstration of shape memory cycle of a 4D-printed SMP microlattice through shape programming under heating, deformation and cooling, and its recovery to original shape under heating (reproduced from Ref. [102] with permission from the Royal Society of Chemistry.); (b) Representation of different programmable hygroscopic deformation modes based on an anti-tetrachiral structure with hygroscopic gradient, shearing and bending deformation modes at 20% RH (left side) and 80% RH (right side) (reproduced from Ref. [104] with permission from the Royal Society of Chemistry.); (c) The demonstration of potential applications of PRMMs such as a free-falling test of the PRMM and a physical illustration of energy dissipation of PRMMs (reproduced from Ref. [108] with permission from the Royal Society of Chemistry.); (d) 3D stair-stepping mechanical metamaterials (SMM) with multiple pathways to zero stiffness, enriched with vibration isolation characteristics include a three-level structure of the SMM in its initial state (state I) and its programmed state (state II), which was achieved either by applying the shape memory effect of SMP or by adding extra mass (adapted from Ref. [109] copyright 2024 Wiley-VCH GmbH); (e) Demonstration of flat panels morphing into foldable structures (the targeted shape), for making them as novel self-expandable structures (adapted from Ref. [110] copyright 2023, Zhejiang University Press); (f) Illustration of the gripper's functional configuration by sequence control through the temporary shape and the whole working process of the robotic gripper (adapted from Ref. [111] under a Creative Commons Attribution 4.0 International License).

activation parameters via multi-physical modelling and a highly nonlinear deploying process, allowing them to be used as reconfigurable robots, as shown in Fig. 6(f).

4. Structure optimization

The relentless pursuit of structure optimization and topology is considered the driving force behind the inevitable growth of mechanical metamaterials in high-end applications [112]. Recently, extensive attention has been directed towards the structural design and optimization of mechanical metamaterials, which include interconnected struts, plates, sheets, or shells with submillimeter to millimeter-scale cavities or pore sizes arranged in repeating units. Computational models of 3D/4D metamaterials are continuously evaluated and refined throughout the design and validation process. Moreover, the integration of complex architectural designs and printing parameters into these computational models is crucial for developing novel mechanical metamaterials with desirable functionalities and satisfactory properties [113]. Numerous lattice geometries can be developed using 3D printing, particularly the FDM-based 3D printing process, which permits the on-demand of multifarious metamaterials with unique internal architectures. This involves optimizing a structure under given constraints, such as excellent stiffness, high thermal dissipation, lightweight, and low cost to achieve the optimal design [114].

To date, various computational tools have been employed to optimize structures for maximizing properties including flexibility, strength, weight reduction and overall performance. In topology optimization, the optimal distribution of material for a specific target is determined by prescribed governing equations and considering initial boundary conditions. Finite element methods are often used to solve the resulting physical problems by iteratively improving a function subject to constraints [115]. The programming implementations of topology optimization include methods such as bidirectional evolutionary structural optimization, solid isotropic material with penalization, and the level set method, etc. [116]. Generally, to optimize an isotropic solid, continuous optimization relaxes the requirement for elements to be either filled or empty, allowing for densities to vary continuously between 0 (empty) to 1 (filled). In contrast, discrete optimization, which can be either randomized or deterministic, begins with an initial guess, generates a new structure, compares the merit of the old and new designs, applies acceptance criteria, and repeats this process until convergence is reached.

Substantial progress has been made in 4D-printed shape-morphing metamaterials, driven by the ongoing evolution of ML technology. ML offers a novel approach to materials design, helping in the establishment of reporting standards and the use of benchmark sets for model comparison and evaluation [117]. Recently, Liu et al. [118] combined ML with high-throughput molecular dynamics (MD) simulation strategies for translating architected materials to the atomistic scale and discovered some atomistic families of disordered mechanical metamaterials. It was revealed by systematic exploration of the forcefield landscape that modest bond directionality promotes the disordered packing of polyhedral, stretching-dominated structures, which are responsible for metamaterial formation (as shown in Fig. 7(a)) between directional and non-directional bonding, including covalent and ionic bonds. This study offered a new insight into a bottom-up atomistic strategy for designing disordered mechanical metamaterials, potentially applicable to conventional upscaled designs. Lim [119] proposed a novel metamaterial composed of rotating trapeziums and triangles. Through a geometrical analysis, the on-axes Poisson's ratio of the metamaterial was formulated. Moreover, by changing the geometrical configuration of the microstructure through internal angles, the effective material properties of the proposed metamaterial showed noticeable changes, including a configuration that enabled the metamaterial to demonstrate a sign-programmable Poisson's ratio. These developed metamaterials are suitable for applications requiring frequent in-situ adjustment of effective properties due to their ease of reconfiguration, tunable mechanical properties, and the ability to program the Poisson's ratio sign, as presented in Fig. 7(b).

ML translates mathematical descriptions of optimal macro/microstructures into a physical 3D structure and generates machine code for printing. Moreover, researchers often use trained ML models to predict the performance of these structures, including mechanical properties and shape morphing under various environmental conditions, by identifying patterns and relationships within the data sets. This approach reduces the need for further experimentation with these metamaterials. For instance, Gao et al. [122] adopted a facile approach to create damage-programmable mechanical metamaterials by adding crack-resisting features and fracture-controlling mechanisms, observed in natural materials into the artificial domain using ML. Through ML designs, damage-programmable cells with advanced toughening functionality, such as crack bowing, deflection, and shielding, were generated more efficiently and optimized for any crack path. Notably, resistance to crack advances and fracture energy increased by up to 1335 %, with the cracks being effectively dissipated at desired locations, compared to metamaterials lacking these mechanisms. This improvement was attributed to the strategic programming of various nature-inspired crack-resisting and dissipation mechanisms, which were activated at various stages of crack propagation. Liu et al. [120] performed S-strut optimization on mechanical metamaterials to investigate compression–torsion conversion (CTC) properties of proposed array-type and cylindrical shell-type structures using finite-element simulations, theoretical calculations, and experiments. Theoretical results indicated that the optimized inclined strut significantly improved the CTC properties of these mechanical metamaterial structures, demonstrating CTC efficiencies exceeding 25%, which is notably higher than those of traditional designs. Moreover, finite-element simulations revealed that the CTC properties of unit cells within 16×5 cylindrical shell-type CTC structures and array-type Z-strut CTC structures exhibited a deformation mechanism similar to that of the inclined strut, as illustrated in Fig. 7(c). This suggests that the CTC efficiency of compression–torsion metamaterials is markedly improved by replacing straight rods with optimized folded struts in cylindrical shell-type and array-type designs. Likewise, Dai et al. [123] designed fiber-reinforced, highly energy-absorbing, and vibration-isolation performance-driven mechanical metamaterials based on triply periodic minimal surfaces through finite element simulations and theoretical analysis. Their study demonstrated that fiber reinforcement significantly improved the energy absorption

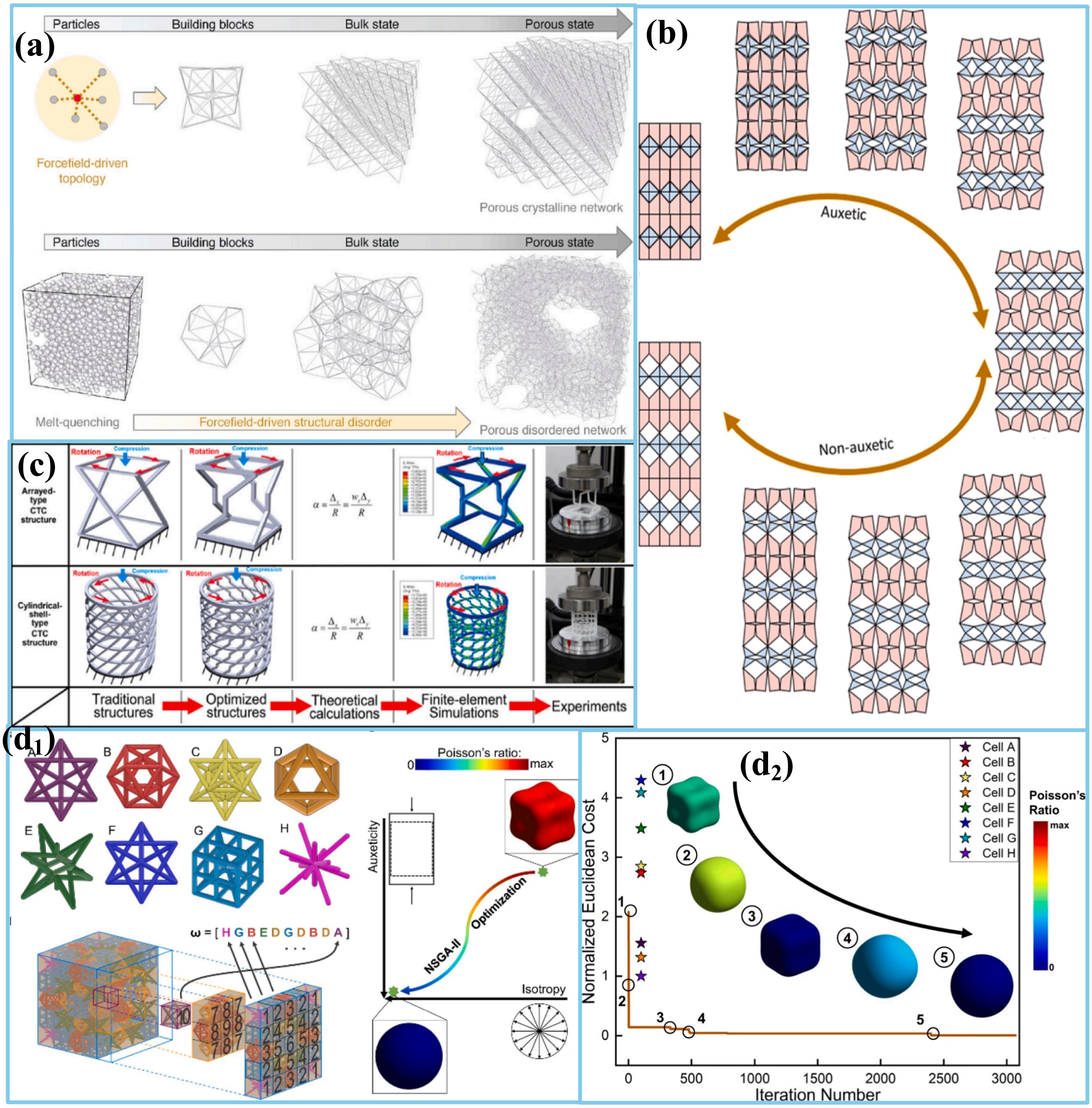

Fig. 7. (a) Representation of fabrication at atomistic scale architecting disordered metamaterials ordered topology which is locally equilibrated by its forcefield, and the pores are randomly introduced into the network to mimic the formation of imperfect crystal which is analogous to conventionally ordered metamaterials from melt quenching (adapted from Ref. [118] under a Creative Commons Attribution 4.0 International License); (b) Demonstration of different configurations of the metamaterial started from its fully-closed state to the states of full extension in horizontal and vertical directions. Each unit cell contains four triangles (denoted in light blue) and four trapeziums (indicated in pink) (adapted from Ref. [119] copyright 2024 Elsevier Masson SAS).; (c) Design strategy of the inclined-strut optimized mechanical metamaterials (adapted from Ref.[120] copyright 2024 Wiley-VCH GmbH); (d_1) Input classification was based on eight types of cubic symmetric input cells, each coded with different colours from A to H. Finally the multi-objective optimization was achieved using an auxetic and isotropic lattice structure, with a stiffness map illustrating the transition. The schematic shows a sphere (bottom left) evolving from an arbitrary shape (top right), indicating perfect isotropy as colours gradually shift from high Poisson's ratio (red) to auxeticity (blue), (d_2) Insets show how structures evolve toward isotropic (perfect sphere shape) and auxetic (blue colour) configurations as iterations increase (adapted from Ref. [121] under a Creative Commons Attribution 4.0 International License). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

and deformation resistance properties of these triply periodic minimal surface structures.

In another novel work by Meier et al. [121] systematically designed highly tunable mechanical metamaterials by integrating modelling, FEA, genetic algorithms, and optimization. The lattice structure controls the stiffness tensor by strategically arranging eight distinct non-isotropic, non-auxiliary unit cell states, as illustrated in Fig. 7(d_1). Additionally, the developed metamaterial lattice structures demonstrated both desired isotropic and auxetic properties due to the optimization of the micro-lattice using the non-dominated sorting genetic algorithm-II, coupled with automated modelling, as presented in Fig. 7(d_2).

Choi et al. [124] designed a self-deformable, soft, and programmable mechanical metamaterial frame using a novel meta-elastomer (ME) substrate with a bidirectional zero Poisson's ratio. The results showed that the unique interaction between the deformation of the MM frame, due to an NPR, and the elastomer matrix, with a Poisson's ratio, led the ME to undergo both structural deformation and length alteration, as depicted in Fig. 8(a_1-a_2). Moreover, the chemically cross-linked ME substrate at the junction interface exhibited exceptional mechanical robustness, sustaining over 180% stretching for more than 10,000 cycles. This study offers valuable insight for developing more stable and reliable stretchable metamaterials for display applications.

Zheng et al. [125] designed novel multifunctional metal metamaterials from Cu-18% Al-l0% Mn-0.3% Si using a laser powder bed fusion. The two unit cell configurations such as S and Ω- unit cell were studied, as highlighted in Fig. 8(b_1). The results showed that the proposed metamaterials exhibited remarkable reversible recovery superelastic strain exceeding 20 % with a 100 % recovery rate and zero Poisson's ratio. Moreover, the 3D-printed metamaterials showed superelasticity, flexibility deformation, one-way, and two-way shape memory effect and multifunctionality, as shown in Fig. 8(b_2).

With the incorporation of bio-inspired structures or mechanisms, the plateau stress of mechanical metamaterials can be improved for effectively improving the energy absorption capability. For instance, Xu et al. [126] studied the compression behaviour and energy absorption capabilities of novel glass-sponge-auxetic structures with concave cells reinforced by diagonal braces, using both compression testing and FEA. The results showed high plateau stress and densification strain, as well as the highest energy absorption and specific energy absorption, attributed to the unique bending-stretching deformation and non-simultaneous fracturing pattern. Moreover, the customizable cell number allowed further optimization of energy absorption ability for specific applications. Likewise, Zhao et al. [127] conducted a detailed numerical simulation procedure to determine the effective macroscopic elastic moduli of cubic bio-inspired metamaterial designs, including gyroid, primitive, and diamond configurations. The simulation results showed that all three metamaterial designs exhibited shear-compliant behavior with a small internal surface thickness and high Poisson's ratio of the base material and compression-compliant material under opposite conditions (low Poisson's ratio and large internal surface thickness). Additionally, the primitive design was found to be highly anisotropic.

5. Shape-memory and energy-absorbing features

Metamaterials have significantly revolutionized the development of lightweight energy-absorbing structures under the evolution of 3D printing. These metamaterials play multifaceted roles, encompassing negative mechanical properties and requiring customized shape-morphing capabilities. Some pioneering studies have demonstrated that metamaterials possess high mechanical resistance, permitting them to withstand a greater number of stress cycles while recovering their shape. For instance, Ren et al. [128] proposed highly customizable and reconfigurable mechanical metamaterials using a PLA/TPU-based multi-material system from FDM-based printing. The unique characteristics of 4D-printed metamaterials, such as customizable and configurable, were evaluated through quasi-static compression mechanical performance, cushion properties, and vibration isolation abilities performed across different material compositions and geometric parameters, as presented in Fig. 9(a). The results revealed that negative stiffness metamaterials exhibited excellent buffering performance due to their enhanced energy absorption capabilities compared to zero stiffness metamaterials. Additionally, negative stiffness metamaterials outperformed zero-stiffness variants in vibration isolation. Moreover, the proposed novel TPU/PLA-based SMP can reprogram the structure of these metamaterials above the glass transition temperature (T_g), enabling unique reconfigurability in terms of structure, stiffness, and properties.

Likewise, Xu et al. studied [129] 4D-printed SMP to demonstrate various cellular metamaterials with negative/zero/positive Poisson's ratio and vibration isolation effects. It was shown that at different stages of deformation, 4D-printed cellular metamaterials with zero Poisson's ratio outperformed cellular metamaterials with negative or positive Poisson's ratio. Moreover, zero Poisson's ratio and positive Poisson's ratio cellular metamaterials exhibited remarkable energy absorption characteristics at temperatures near to Tg whereas negative Poisson's ratio and positive Poisson's ratio cellular metamaterials showed greater absorption capacity at lower temperatures. Interestingly, all cellular metamaterials had remarkable shape recovery rates (>86.67 %) as shown in Fig. 9(b). The proposed research has a promising role in designing various architecture for, automotive engineering, innovative wings biomedicine and nanotechnology.

In another novel work, Wan et al. [130] designed Tachi-Miura Polyhedron (TMP) origami metamaterials having reconfigurable shapes and programmable mechanical performance from 4D printing. A multi-branch modelling approach was adopted to characterize the SMP for illustrating their complex mechanical responses around its T_g. Results showed that the TMP origami metamaterials can be programmed into various folded shapes as presented in Fig. 9(c_1 and c_2) with different mechanical properties. Moreover, TMP origami structures exhibited various deformation modes by switching between monostable and bistable states and achieved NPRs, under proposed SMEs, under programming and controlling temperature. The proposed 4D-printed origami metamaterials have a prominent role in energy absorption devices, self-deployable structures, and the flexible electronics field. Li et al. [131] proposed an auxetic mechanics metamaterial from 4D printing with shape memory capability for demonstrating NPR and energy absorption abilities. Insights of this study showed that during in-plane compression the bending deformation was the dominant deformation mechanism, thus, preventing stress oscillation and later applied as an energy absorber. In addition to that the programmable and reconfigurable features of proposed mechanical metamaterials were determined utilizing a hot water bath. Particularly when subjected to a vertical tensile or compression displacement along the Y-direction to program it into a temporary configuration under hot water. This allows the specimen to maintain the external force which was later cooled to room temperature, and the external force was released to make it maintain a temporary configuration, as presented in Fig. 9(d).

Liu et al. [132] proposed a facile way for fabricating curved porous silicone foams with both tailored *meta*-mechanical properties and 4D-printed bending curvature by a DIW of composite silicone inks embedded with thermally expandable microspheres as foaming agents. Results demonstrated that the printed bilayer structures demonstrated shape-shifting behavior by utilizing the strain mismatch under the thermal stimulus, as illustrated in Fig. 10(a). Moreover, under compression, silicones containing stacked bilayer filaments showed negative stiffness, resulting in improved energy absorption capacity. In addition to that using different printing structural designs, it can be fine-tuned to dissipate energy and absorb shock while protecting objects with curved shapes. Likewise, Li et al. [133] investigated dynamic biomimetic mechanical metamaterials of the dome structure array developed by FFF-based printing. The dynamic transformation of 2D planar structures into 3D structures was derived from the thermal stimulation treatment along with the programming of the designed structural modes. Results showed that energy absorption characteristics were improved by the number of circular units. Also, circular units with different diameters yield deformation rises of various heights under thermal stimulation, as shown in Fig. 10(b_1–b_3), making them an ideal fit for complex surfaces which are suitable for special applications for strengthening and cushioning of numerous weapons, vehicles, launch structures, and heavy equipment.

Lightweight, and high-strength 4D-printed metamaterials with energy-absorbing structures are extensively employed in many fields, such as automotive, aerospace, medical, sports protective equipment, and flexible electronics, where the material and structure are the two leading factors [134]. Zhao et al. [135] investigated ultralight, high strength, high specific energy absorption efficiency and stretching-dominated metamaterial using FDM. The results showed that 3D-printed bionic-designed shape-memory (e.g. PLA/Fe_3O_4), materials

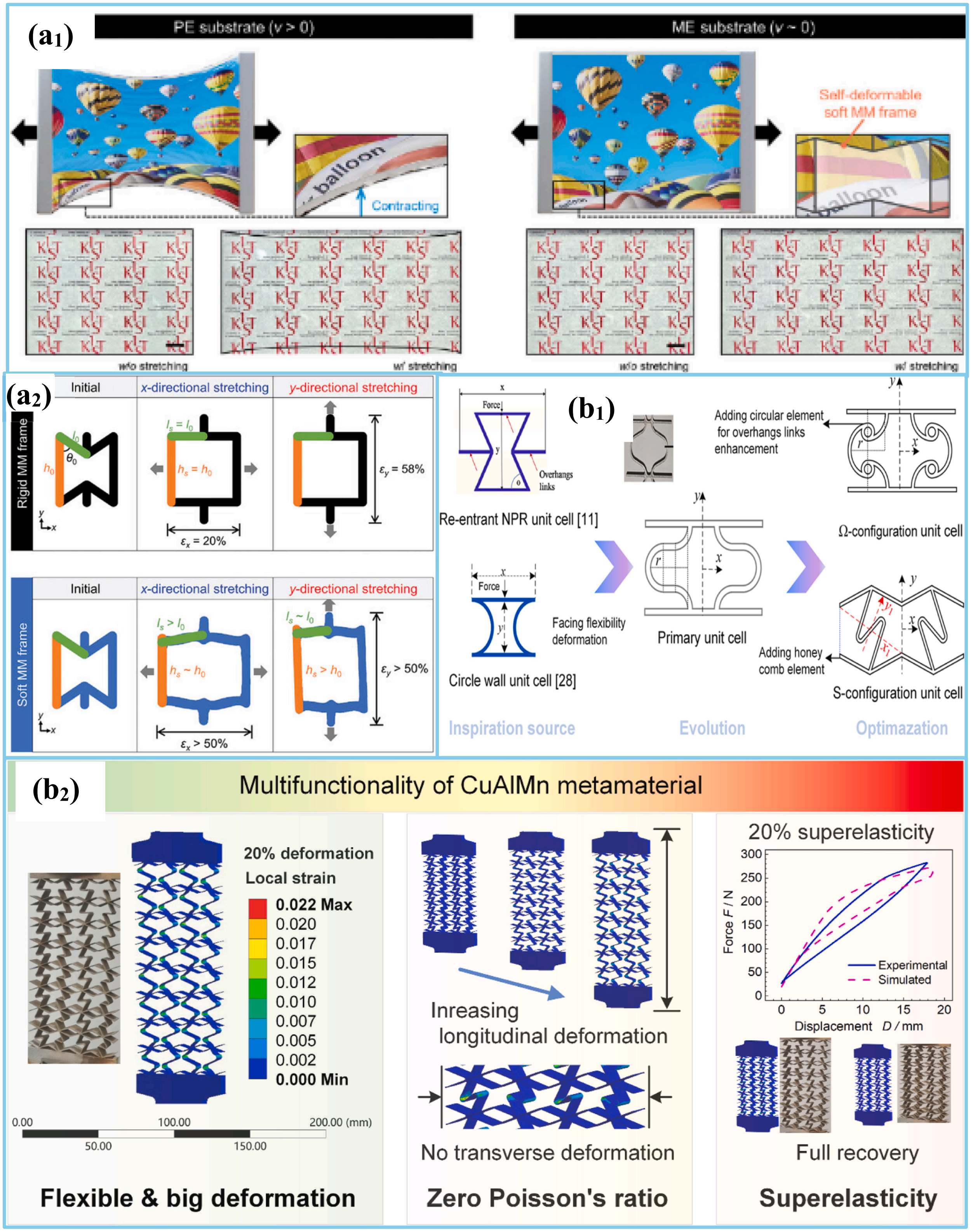

Fig. 8. (a_1) Self-deformable soft metamaterials for bidirectional zero Poisson ratio substrates in stretchable displays and demonstrate large axial stretching of pristine elastomer (PE) (positive Poisson ratio) and ME (zero Poisson ratio) substrates and (a_2) illustration of the unit cell deformation patterns of rigid and soft metamaterial frames under bidirectional stretching along the x–y axes (adapted from Ref. [124] under a Creative Commons Attribution 4.0 International License); (b_1) Demonstration of novel metamaterial unit cell designs such as the inspiration source, the primary unit cells, and finally the optimization process; (b_2) The multifunctional performance of metamaterials demonstrating including superior flexibility deformation, zero Poisson's ratio and superelasticity (adapted from Ref. [125] copyright 2024 Elsevier Ltd.).

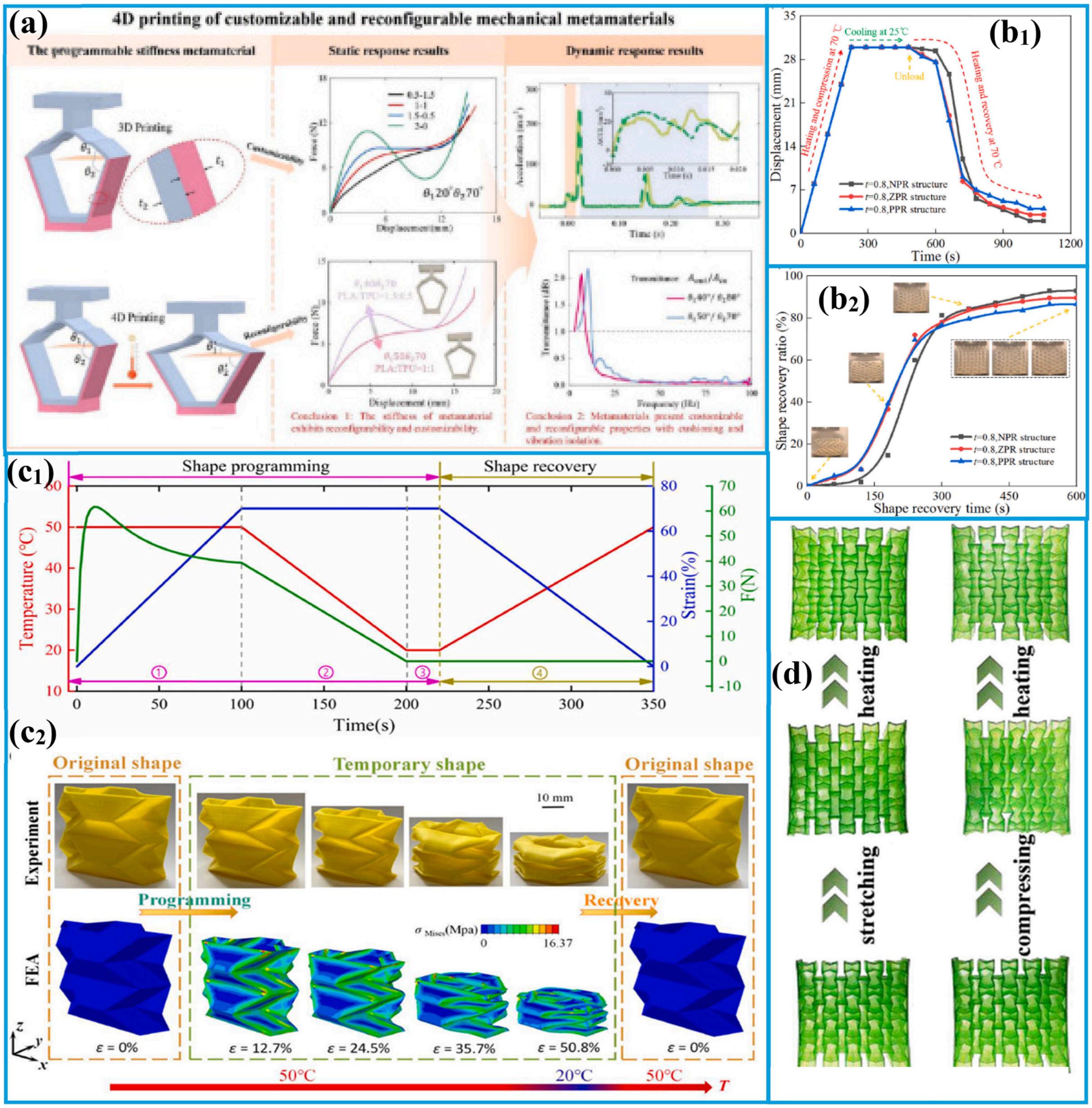

Fig. 9. (a) The demonstration of the shape memory process representing the reconfiguration process under static response in which static mechanical properties of printed metamaterials reconfiguring from positive stiffness to zero stiffness and from zero stiffness to positive stiffness while printed metamaterial customizability and reconfigurability were evaluated through vibration isolation performance and cushioning under dynamic response (adapted from Ref. [128] copyright 2024 Elsevier Ltd.); (b_1−b_2) 4D-printed cellular metamaterials shape memory cycle representing (b_1) Time-displacement curve from which three cellular metamaterials were heated at 70°C and compressed 30 mm at the same loading rate in the first 480 s after that temperature is uniformly lowered from 70°C to ambient temperature under the cooling stage, (b_2) Graph of recovery time-shape recovery rate. The structure's shape recovers from the 480 s to the 1080 s in (b_1), and as the temperature rises from 25°C to 70°C, the cellular metamaterial begins to return to its original shape due to the glass-rubber transition. This stimulates the shape memory effect of the cellular metamaterial (adapted from Ref. [129] copyright 2023 Elsevier Ltd.); (c_1-c_2) TMP origami metamaterials during compression demonstrating shape memory behaviour (c_1) Thermomechanical cycle graphs, (c_2) Simulation and experimental results for shape-memory behaviour (adapted from Ref. [130] copyright 2023 Elsevier Ltd.); (d) Description of the programmable and reconfigurable performance of the metamaterials includes processes such as stretching followed by heating recovery process, and compressing followed by heating recovery process (adapted from Ref. [131] copyright 2024 Elsevier Ltd.).

demonstrated excellent shape-memory behavior following four stages, as illustrated in Fig. 10(c). The first stage consisted of heating to 90°C and maintaining it for five minutes to reach thermal equilibrium. A 50 % deformation was then applied to the metamaterials. The third stage involved cooling the temperature from 90°C to 20°C at a rate of 2.5°C per minute. Afterwards, the external force was unloaded while the deformation was retained. During the last stage, the temperature was reheated from 20°C to 90°C at a rate of 2.5°C/minute. As the temperature increased, the metamaterial gradually returned to its original configuration. This study sheds new light on the shape-memory function of bionic materials for developing novel tissue stents.

With the development of biomaterial-based AM technology, metamaterials have revolutionized the design of energy-absorbing structures. This advancement enables the creation of better energy-absorbing structures using novel biomaterials, which will contribute to the production of cost-effective structures for many sustainable applications. Recently, Zhang et al. [136] developed novel reusable metamaterials using PLA and polyethylene terephthalate glycol (PETG)-based SMP

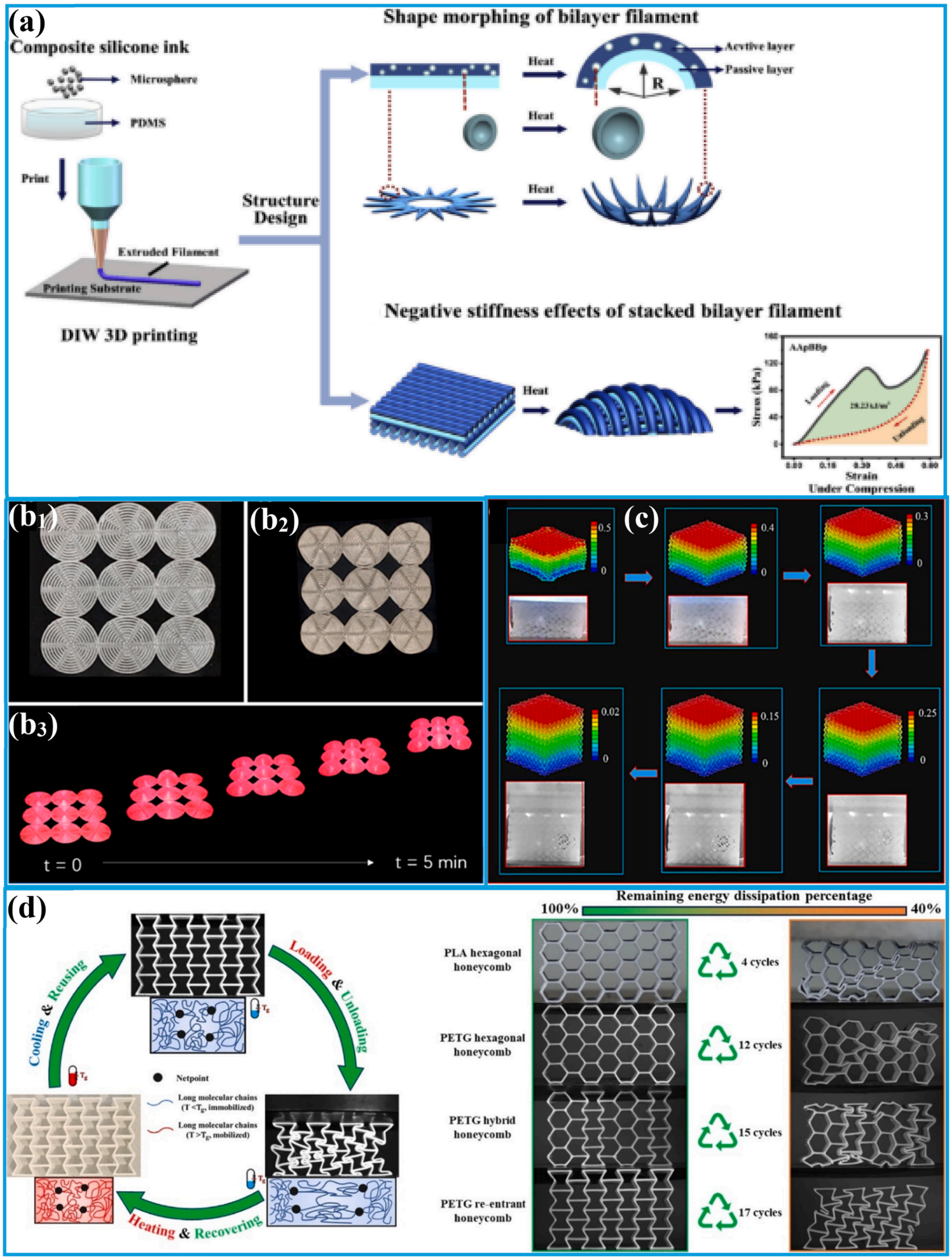

Fig. 10. (a) Demonstration of shape morphing of DIW-printed structures with bilayer filament and negative stiffness effect of typical structures under compression with stacked bilayer filaments (adapted from Ref. [132] copyright 2024 Elsevier Ltd.); (b_1-b_3) Thermal stimulation process for 3D-printed samples, (b_1) 3D-printed flat structure, (b_2) Structure after thermal stimulation, (b_3) Representation of deformation process during heat treatment (adapted from Ref. [133] under a Creative Commons Attribution 4.0 International License); (c) Demonstration of shape-transformation of lattice metamaterials, comparing simulation and experiment results (adapted from Ref. [135] copyright 2022 Elsevier Ltd.); (d) Representation of SME under thermal stimulation, where T_g is linked with phase changes, and energy dissipation under multiple cyclic loading is shown as a percentage for different PETG-based structures (adapted from Ref. [136] under a Creative Commons Attribution 4.0 International License).

from FDM-based printing. Their reusability was studied through various mechanisms such as the effect of printing materials, under cyclic programming and unit-cell types on mechanical degradation. Insights of this study showed that the PLA hexagonal honeycomb dissipated 22% more energy in comparison to PETG under one single compression cycle (Fig. 10(d)). This is attributed to PLA's higher elastic modulus leading to a larger critical buckling load for segments in honeycomb structures. On the other hand, the PETG re-entrant honeycomb dissipation more energy (25%) due to its NPR as well as uniform deformation pattern than the hexagonal counterpart. Moreover, as a result of the better ductility, PETG hexagonal honeycomb under multiple compression cycles particularly at cycle 6 maintained an energy dissipation capacity of 78.3%, which is almost 3.5 times as compared to PLA structures.

Zhao et al. [137] introduced four different types of 3D chiral and anti-chiral metamaterials: original chiral, diagonal symmetry chiral metamaterials, diagonal symmetry anti-chiral and central symmetry anti-chiral metamaterials, all produced through 3D printing. The reported results indicated that these metamaterials showed tension-twist coupling behaviour in chiral metamaterials. Moreover, their significant deformation ability and shape-memory behaviour, as shown in Fig. 11(a_1–a_3), combined with programmatic calculations and distributions, highlight their potential as novel mechanical metamaterials for

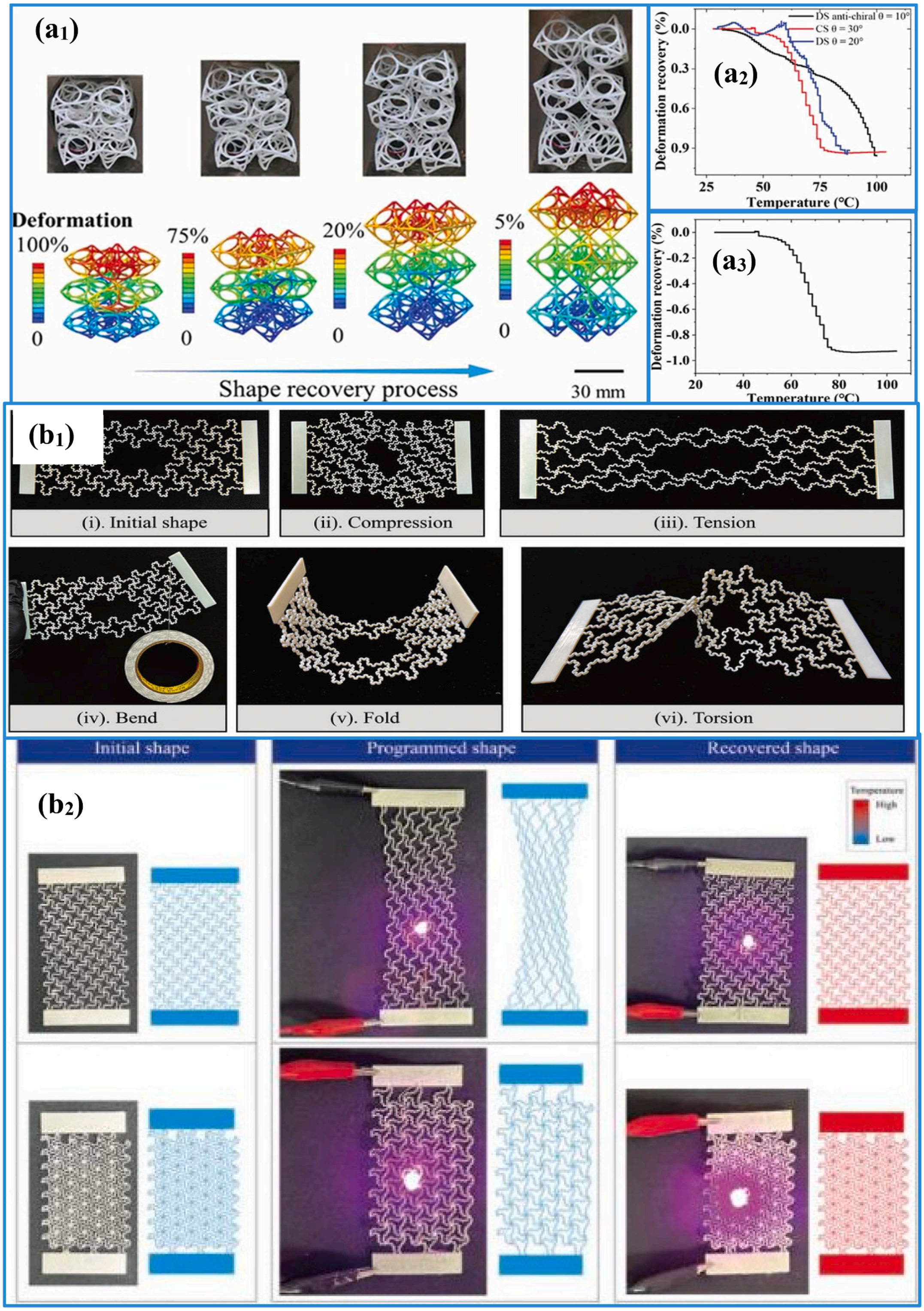

Fig. 11. (a_1- a_3) Representation of shape recovery performance of the metamaterials includes: (a_1) A comparison between simulation and experiment results, (a_2) Graph for deformation recovery vs. temperature and (a_3) Metamaterials configuration at various temperatures during the shape recovery process (adapted from Ref. [137] under a Creative Commons Attribution 4.0 International License); (b_1) Demonstration of five programmed states such as compression, tension, bend, fold and torsion, (b_2) Illustration of LED devices based on fractal metamaterials (FEM results also shown for comparison) such as rectangular and triangular with shape reconfigurability and conductive in various states (adapted from Ref. [138] copyright 2022 Wiley-VCH GmbH).

numerous engineering applications. Likewise, Wang et al. [138] investigated novel 3D-printed fractal metamaterials that demonstrate bionic stress–strain matching, conductivity, lightweight, imperfection and insensitivity properties. Insights of this study showed that these fractal metamaterials showed remarkable tensile performance. Moreover, due to the wide design space of the fractal metamaterials and the four types of imperfect fractal metamaterials, the J-shaped stress–strain curves of different biological tissues and organs are excellently matched. Lastly, the fractal metamaterials showed a reconfigurable shape and the ability to adapt to the external environment under the thermal stimulus, as presented in Fig. 11(b_1 and b_2), due to the shape-memory effect of the Vero material. This work highlights the realization of fractal metamaterials in creating multifunctional soft devices.

Zhang et al. [139] investigated the role of 4D printing of SMPs in nanophononics in the submicron length scale. By using TPP lithography, the SMP photoresist based on VeroClear produces print features at a resolution of 300 nm half pitch. It was shown that both optical and microtopography properties of the structures can recover well attributed to excellent shape memory effect at the submicron scale, as shown in Fig. 12(a). The proposed high-resolution printing with tunable reversibility holds great promise for a wide range of novel applications as information hiding for anti-counterfeiting, temperature-sensitive labels, and tunable photonic devices. Gisario et al. [140] studied the FDM-based 4D-printed behaviour of six chiral structures regarding their mechanical deformation behavior under quasi-static compressive loading and shape recovery performance using PLA as SMP, as shown in Fig. 12 (b). The results showed that the density of the model has a strong influence on the energy absorption and the degree of damage increases with an increasing number of load cycles. The lozenge grid is considered the strongest and most reliable geometry as no damage was observed while increasing compression cycles. In addition to improving mechanical strength and absorption energy, the increase in grid density also lowers the microcracks in the geometry itself due to repeated load cycles. Desole et al. [141] evaluated the impact energy performance and subsequent shape-memory of solid cellular structures (strut-based, triply periodic minimal surfaces and spinoidal) using PLA as SMP. Three categories of impact tests such as high, medium and low impact energy were characterized from these structures. Insights of this study showed that triply periodic minimal surfaces demonstrated high and medium impact energies, due to the existence of few internal defects. Fig. 12(c_1) highlights the comparison between the two structures tested at high impact energies while Fig. 12(c_2) associates the results for five various specimens when the structures reach a status of permanent failure as the impact energy varies. Spinoidal structures showed low-impact energy performance due to their geometric characteristics. Finally, after the structures were deformed, they were immersed in a thermostat bath at 70-°C, a temperature higher than the glass transition of PLA, which allowed them to recover their shape.

6. Applications of 4D-printed metamaterials

Metamaterials combined with 4D printing are drawing great attraction worldwide and have the potential to replace conventional materials. This burgeoning technology can be applied in myriad technological fields including, but not limited to tissue engineering, intelligent robotics, biomedical devices, drug delivery systems, origami structures, wearable devices, construction, and aerospace [142]. This section reviews both functional and structural applications of 4D-printed mechanical metamaterials. Structural applications rely on their NPR, and energy absorption properties whereas functional applications are based on their design flexibility, shape morphing capability, and optical, electrical, thermal, magnetic, and biomedical properties.

6.1. Smart actuators

4D-printed metamaterials have been vastly applied to develop different soft robots such as soft controllers, sensors, and soft actuators, using electro-active polymers, magneto-active polymers, light-driven polymers, among others. These advancements allow the replacement of traditional rigid-bodied robots [143]. Shape-shifting metamaterials are considered a powerful tool for developing reconfigurable smart actuators [144]. It is expected that this tool will not only change their shape but also induce significant changes in their material properties upon activation. Recently, Wu et al. [145] studied compression-twist coupling building blocks embedded with chiral features as distinct mechanical metamaterials from multimaterial DLP-based 4D printing. By mechanically compressing each chiral unit at a selected high temperature, its temporary shape can be maintained when the load is removed at a reduced temperature. Insights from this study showed that by harnessing the cooperative deformation of spatially heterogenous tessellation that undergoes local expansion or shrinkage, and relying on encoded Poisson response, highly tunable Poisson's ratio and shape-morphing metastructures were developed. Moreover, these chiral metastructures, with unique morphing modes, were employed to construct an untethered multimodal soft gripper capable of unscrewing a bottle cap, as shown in Fig. 13(a), due to the programmed functionality at the module level. This finding suggested that the developed modular chiral metamaterials with embedded chirality and flexible deployability, are promising candidates for functional soft machines as well as active shape-morphing structures. Likewise, Jiao et al. [146] created biomimetic bouquets based on origami metamaterials that store and transmit information in three dimensions: quantity, species, and colour. The deformation for the origami metamaterial unit configurations is presented in Fig. 13(b_1). Additionally, biomimetic bouquets allowed floriography to be expanded into the fourth dimension of status, which significantly reduced the amount of information required via the traditional three dimensions. Based on the crease patterns of units and their overall structures, the origami metamaterials mimicked the flower's growing process (buds and blooms), abstracted into the numbers 1 and 0, as presented in Fig. 13(b_2). This study highlighted the important aspect of designing and assembling the new-generation origami mechanical metamaterials for next-generation bio-inspired intelligent devices and information systems. Li et al. [147] showed that by using 4D-printed fractal-inspired metamaterials with microlevel biomimetic electrodes, this sensor offers both functional and structural reconfiguration. The results showed that the cylindrical microarrays on the samples' surface returned to their original shape when the samples were reheated above the Tg (80-°C) (as shown in Fig. 13(c) on the left side) during out-of-plane testing. Moreover, the SEM images of micro-columnar SMP arrays with a spacing of 10 μm during the out-of-plane shape memory cycle. The in-plane SME circle tests are illustrated in Fig. 13(c) on the right side. These unique features of the proposed sensor along with their reconfiguration ability in mechanical and electrical properties under stimulant environments and excellent sensitivity to signal feedback with superior flexibility make them unique 4D-printed reconfigurable sensors compliance to the human body and have broad applications in the wearable electronics field.

Dezaki and Bodaghi [148] introduced meta-laminar jamming (MLJ) actuators using polyurethane SMP-based meta-structures, such as circles, rectangles, diamonds, and auxetic shapes, through 4D printing. These MLJ actuators performed as soft or hard robots, with their performance through hot and cold programming triggered by negative air pressure. The results showed that 4D-printed MLJ actuators with auxetic meta-structure cores demonstrated superior contraction and bending performance along with remarkable shape recovery (100%) after stimulation. Additionally, various shape-locking features, such as zero-power holding and gripping enabled the actuators to support a 200 g weight without any power input. This facile work provides valuable insights into the designing of metaactuator-based gripping devices for a wide range of engineering applications.

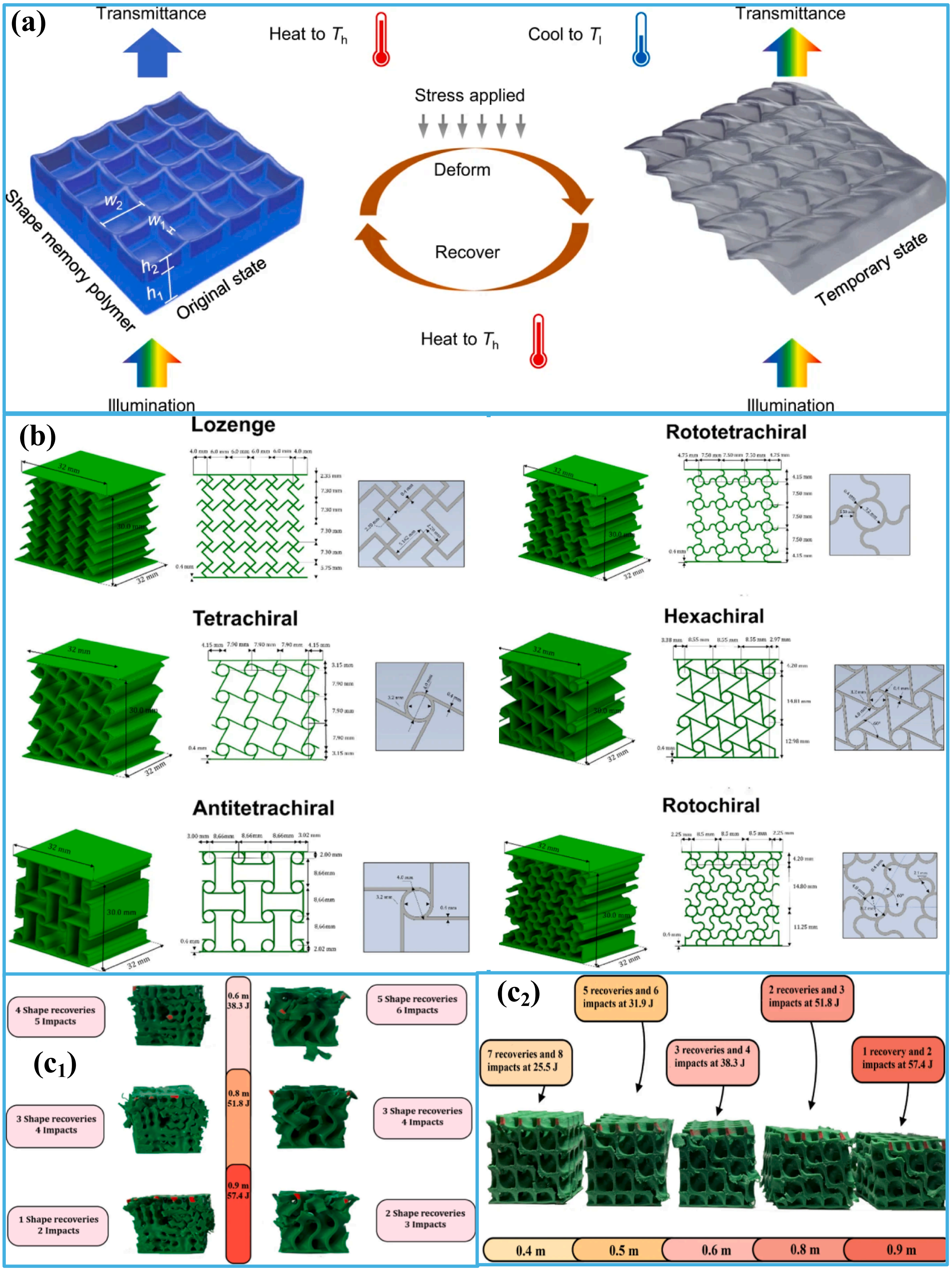

Fig. 12. (a) As-printed structures that transmit only visible light with limited wavelength range due to upright grids (left) function as a structural colour filter, next is structures deformation at high temperature flattens the nanostructures (right) interpreting it colourless, where it remains in an invisible state after cooling to room temperature and finally a demonstration of 4D printing (shape morphing behvaiour) where heating recovers both colour of nanostructures and the original geometry at submicron level (adapted from Ref. [139] under a Creative Commons Attribution 4.0 International License); (b) Demonstration of energy absorbing 4D printed *meta*-sandwich structures: load cycles and shape recovery (adapted from Ref. [140] under a Creative Commons Attribution 4.0 International License); (c_1-c_2) Comparison of various gyroid and cubic Spinodal structure for various energy levels and subsequent shape recovery performance, (c_2) Diamond structure at different energy levels and impact heights (adapted from Ref. [141] under a Creative Commons Attribution 4.0 International License).

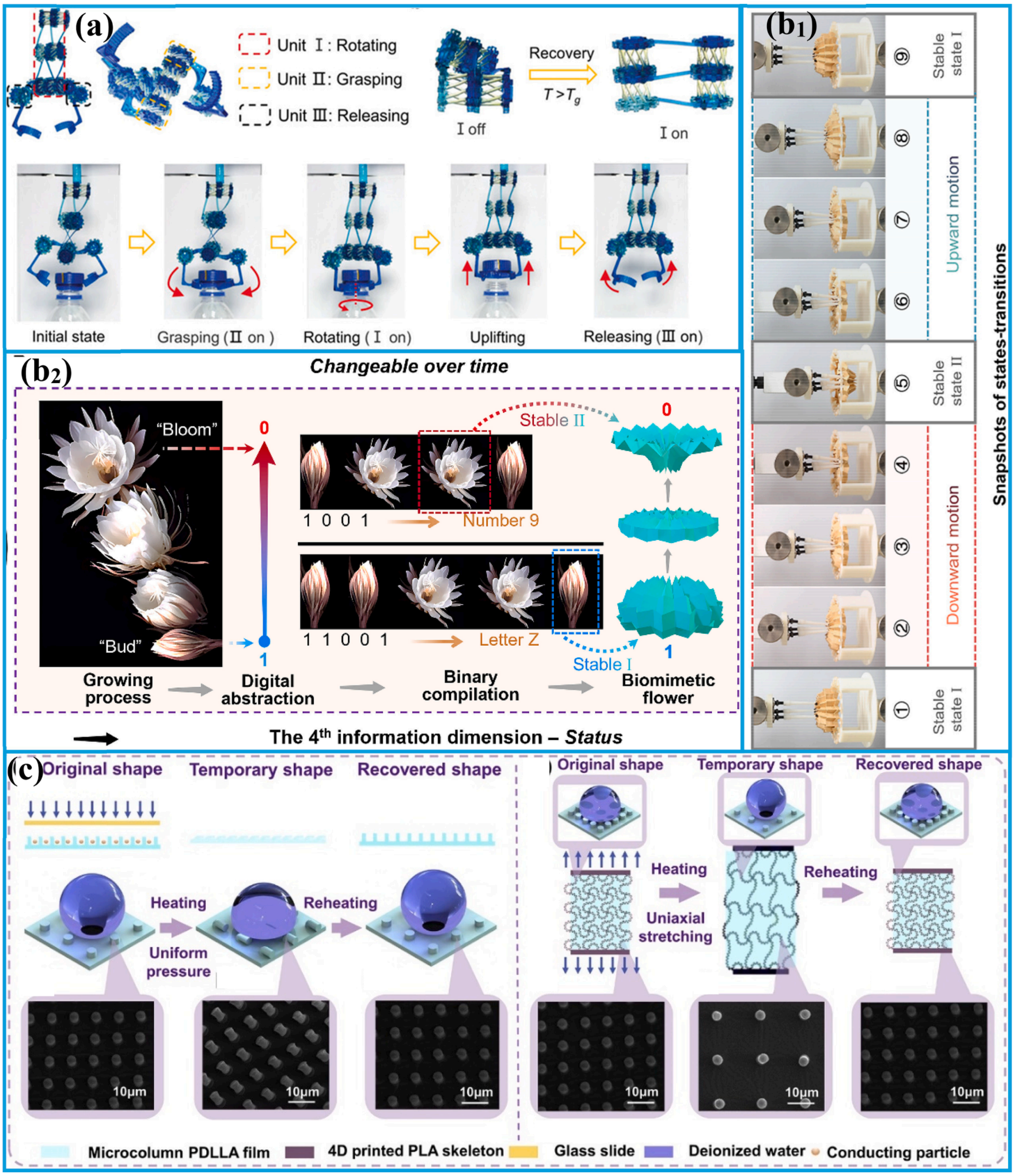

Fig. 13. (a) Demonstration of performance such as rotating, grasping, and releasing units soft actuator under thermal actuation and untethered multimodal soft gripper to unscrew a bottle cap under sequential actuation (adapted from Ref. [145] copyright 2023 Wiley-VCH GmbH); (b_1) Photos of the origami metamaterial unit's deformation configurations, where 1 and 5 are symmetric stable states, (b_2) Representation of binary digit abstraction of main performance in the lily's growing process including bud and bloom over time, information storage and expression instances, such as number 9 and letter Z, and finally illustration of the lily-inspired origami metamaterials changeable over time unit with bistable configurations (adapted from Ref. [146] copyright 2024 the Author(s) Published by Elsevier Inc.); (c) Representation shape memory circle for cylindrical microarrays on the samples' surface returned to their original shape for both in-plane and out-plane testing (adapted from Ref. [147] copyright 2022 Elsevier Inc.).

6.2. Biomedical areas

4D-printed metamaterials have also become a game-changer for the biomedical sector and have been extensively explored to develop biomedical devices [149], as illustrated in Fig. 14. This technology provides innovative solutions by developing patient-specific smart products and dynamic tissue constructs for regeneration, as well as helping in performing minimally invasive procedures [150]. It also helps to develop dynamic tissue constructs, allowing the printed architectures to biologically mimic the properties of native tissues. The self-deformation property of 4D-printed metamaterials is highly beneficial for treating orthopaedic abnormalities. Furthermore, these models can alter their properties when exposed to an external stimulus [151].

4D-printed biorobots can help to deliver therapeutic agents and drugs [153]. Moreover, nano-sized medical devices like bio-actuators and biosensors can monitor the changes in tissues and cells [154]. Similarly, the shape morphing and self-assembly features of 4D-printed materials have been explored by researchers, making them highly suitable for developing bio-sensors, stents, and artificial organs. For instance, origami-derived biomedical devices developed through 4D printing with self-assembly properties can be implanted into the human body through minimally invasive procedures [155]. Recently, van

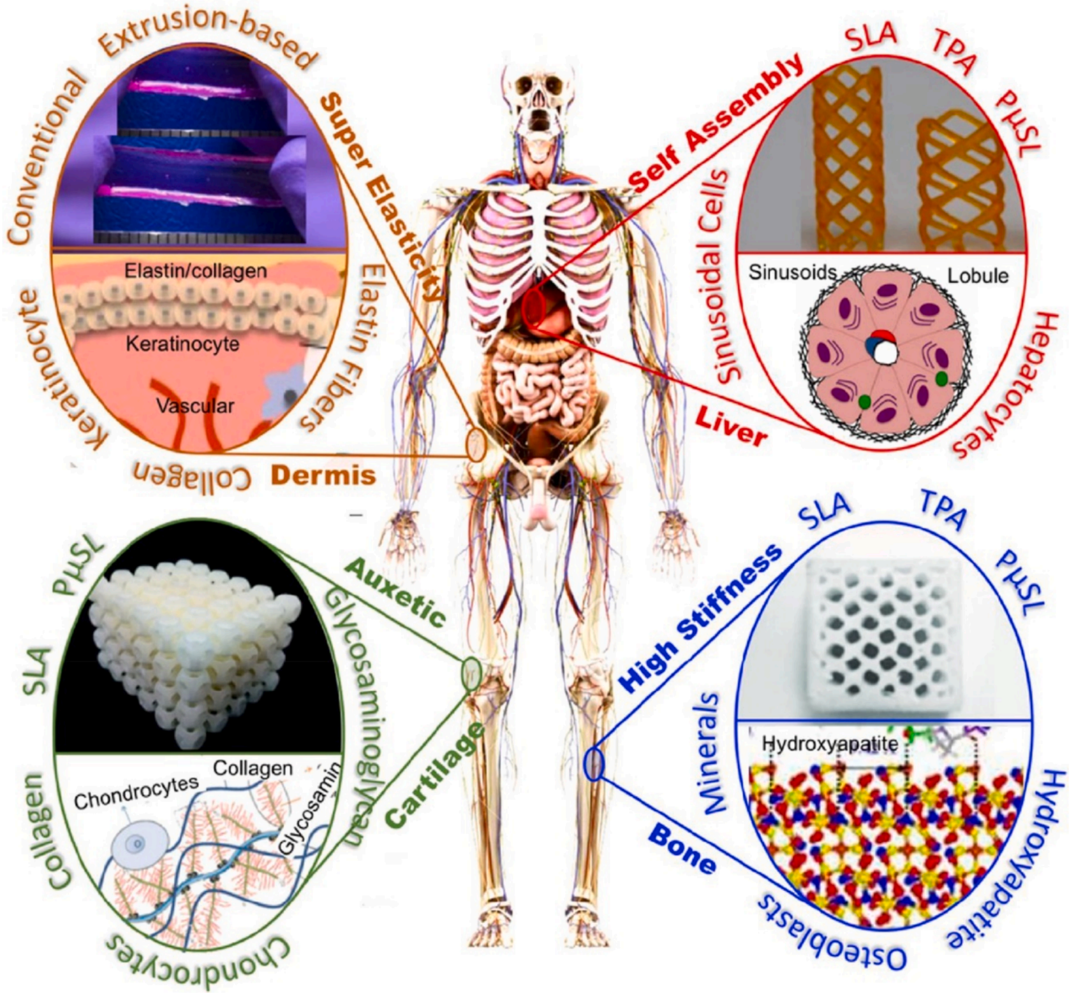

Fig. 14. Novel models for meta-biomaterials and their implementation in biomedical applications for various four organs in tissue engineering and regenerative medicine, such as auxetic models for articular cartilage, super-elastic models for dermis/skin tissue, self-assembly models for liver, and high-stiff models for cortical bone tissue (adapted from Ref. [152] copyright 2020 Elsevier Ltd.).

Mansen et al. [156] adopted a facile approach for the fabrication and programming of 3D-to-3D shape-changing materials using PLA through FDM. The study demonstrated both in-plane and on-curvature anisotropies of these 4D-printed constructs were achieved as a result of printing the specimens on a rotating shaft. Furthermore, a rational design for the shape-shifting behavior was employed in two designs, involving stiff rings connected by flexible elements, to change properties, including stiffness and Poisson's ratio. Furthermore, the proposed reconfigurable materials can be employed as deployable medical devices, including bifurcation stents, as shown in Fig. 15(a_1 and a_2).

4D-printed metamaterials featuring creative design concepts such as kirigami, origami, or auxetic structures open new horizons for fabricating vascular stents with improved crimping-expansion behavior and conformability. These novel vascular stents are anticipated to revolutionize the treatment of vascular diseases in the near future [157]. For instance, Xin et al. [158] developed novel chiral metamaterials with tunable, programmable, and reconfigurable properties using PLA from 4D printing. Their results demonstrated a transition in metamaterial deformation from bending-dominated to stretching-dominated with an increasing λ, leading to auxetic behavior and a nonlinear "J"-shaped σ–λ behavior with a maximum strain of 90%. Moreover, these developed 4D-printed auxetic metamaterials demonstrated successful shape memory programming (as shown in Fig. 15(b_1), with tunable, and reconfigurable mechanical properties that match specific tissues/organs (i.e., pig belly skin, iliac artery, muscle fibers, and dog lungs) and can convert between two biomaterials. Furthermore, the proposed materials are suitable for use in flexible electronics, including LED-integrated devices, as presented in Fig. 15(b_2).

6.3. Self-deployable structures

Nature provides numerous examples of self-deploying structures, such as the wings of earwig and the flaps of peacock spiders. For instance, earwigs have self-deployable wings that enable them to cover large areas and negotiate tight underground spaces. In the realm of 4D printing technology, self-deployable trusses, lenticular tubes, and hinges are notable self-assembled structures [159]. Similarly, deployable antennas and solar cells for spacecraft and satellites, developed through 4D-printed metamaterials, hold huge potential in space programs. These metamaterials are expected to offer high energy efficiency, adaptability, and multifunctionality, paving the way for innovative advancements in art, engineering and various fields, including robotics, architecture, consumer products, and space technologies [160]. Recently, Yan et al. [161] introduced an effective design strategy for fabricating reversibly self-deployable metamaterials inspired by push puppets, using SLA-based printing. Their study demonstrated that particle jamming in the beads permitted for dynamic tuning of the actuator's mechanical properties while maintaining minimal structural changes. Additionally, 3D-printed metamaterials demonstrated significant tunability in bending-dominated configurations after deployment, with over a 50 %

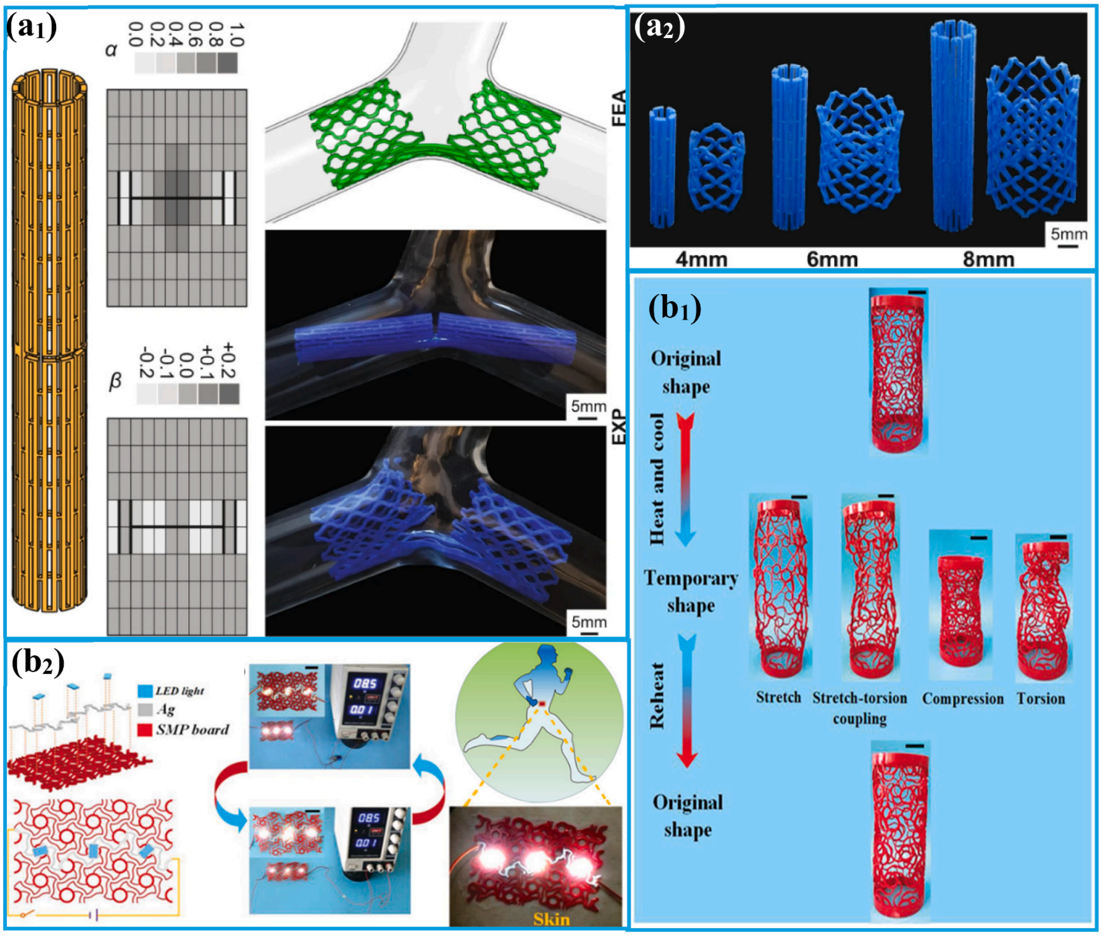

Fig. 15. (a_1) Bifurcation stent deployment within a artery model, (a_2) The deployable stents dimensional scaling (adapted from Ref. [156] under a Creative Commons Attribution 4.0 International License); (b_1) Representation of shape memory programming process of the cylindrical shell (scale bar is 30 mm); (b_2) Demonstration of potential application of LED integrated devices with metamaterials, including the exploded side and circuit diagram, the optical snapshot before and after programming, and the device on the skin (scale bar is 20 mm) (adapted from Ref. [158] copyright 2020 Wiley-VCH GmbH).

change in damping capability and up to 35 times increased stiffness, as presented in Fig. 16(a). This research advances the design of lightweight, reversibly self-deployable, and tunable metamaterials for a wide range of applications in reconfigurable architectures, soft robotics, and space engineering.

The shape-morphing ability of metamaterials with precise control over deformations has attracted considerable attention for various innovative applications [162]. For instance, Liu et al. [163] proposed a highly responsive chiral mechanical metamaterial using prestressed bistable metallic shells. This metamaterial, designed with an anti-chiral arrangement of slender bistable shells, can conform to connected cylindrical cores, allowing a tunable morphing amplitude that can theoretically extend to infinity. Their results showed that the developed smart trapper, based on these metamaterials, can capture moving objects and function as a phononic structure with adjustable band gaps, as presented in Fig. 16(b_1). Moreover, the bistable shell with a transitional speed of 7.56 m s^{-1} transforms from the extended state to a rolled-up state, which offers the 2D and 3D metamaterials with 25.38 and 101.14 times body area/volume variation per second, respectively, as presented in Fig. 16(b_2). This research enhances the understanding of designing novel metamaterials with ultrafast and large-amplitude shape-reconfigurability. Li et al. [164] created compact origami metastructures based on polyhedra using hierarchical closed-loop mechanisms. This approach for intricate geometric constraints reduces the number of active degrees of freedom required for effective shape-morphing behavior, achieving almost 10^3 architectural configurations. These transformable hierarchical structures have the potential for use in self-reconfigurable and self-deployable architectures, offering excellent scalability and multidirectional locomotion. As proof of concept, the developed hierarchical origami metastructures can be employed in fast self-deployable and self-reconfigurable robotics, as well as multi-task reconfigurable space robots, as presented in Fig. 16(c), for transformable buildings and infrastructure.

Ye et al. [165] developed multimaterial thick-panel origami structures using PLA/TPU by printing directly through a single FDM multimaterial 3D printer, resulting in a 3D self-locking thick-panel origami structure. These self-locking Miura-origami units, as shown in Fig. 16 (d), were foldable and could withstand compression while converting vertical compressive forces into horizontal tensile forces through an unconventional push-to-pull deformation mode. The structure demonstrated durability by sustaining over 100 cycles of 40 % compressive strain and supporting loads exceeding 11,000 times its weight. Likewise, Zhao et al. [166] introduced origami-inspired 4D-printed metamaterials with programmable stiffness, adjustable through changes in geometric parameters and configurations due to their shape memory properties. The results highlighted significant improvements in shape reconfigurability, mechanical programmability, and adaptability, with the stiffness of the structure being tunable. Furthermore, the integration of triboelectric nanogenerator gait monitoring exemplifies the potential of 4D-printed shape memory metamaterials for intelligent monitoring and programmable shock absorption.

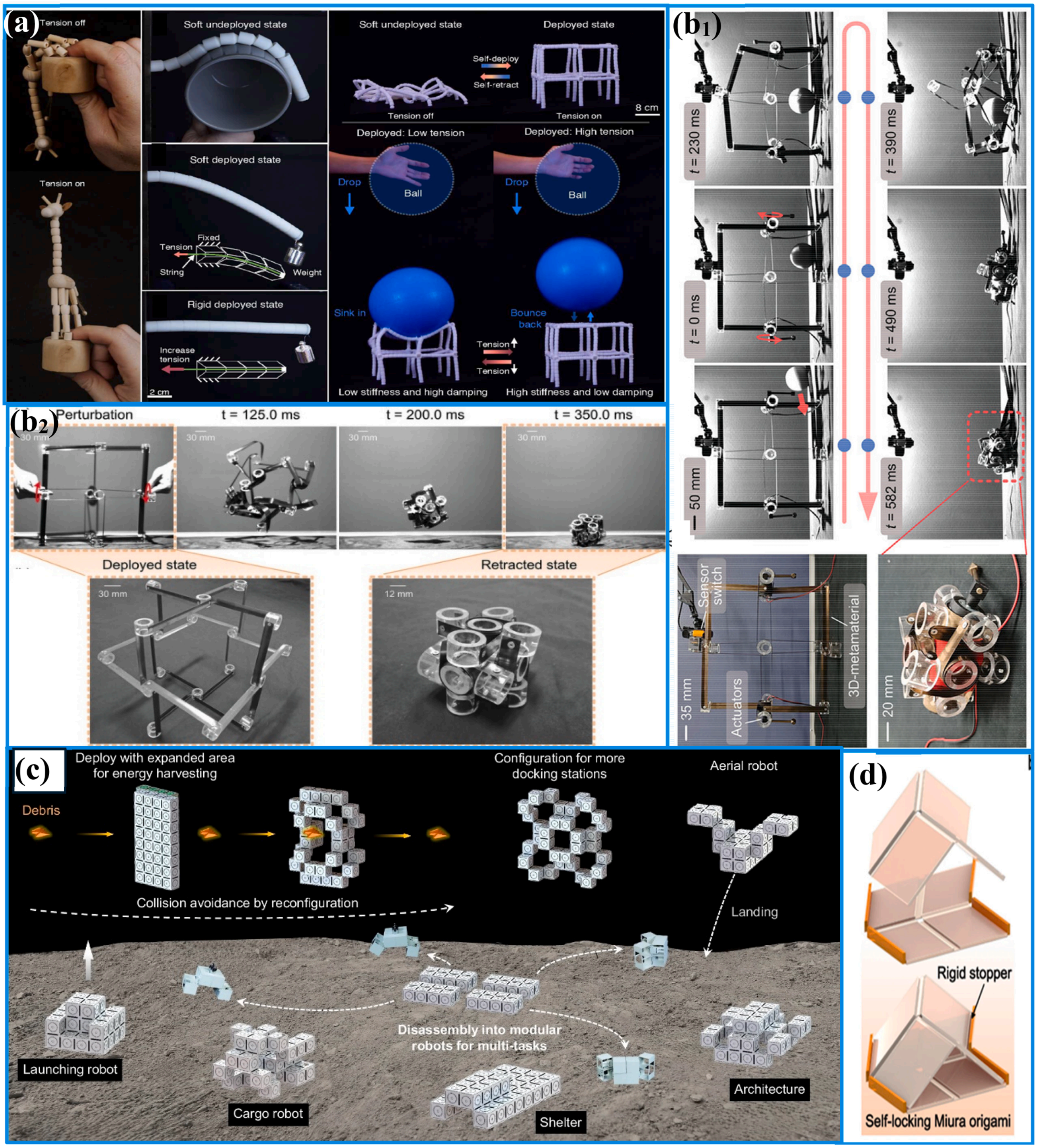

Fig. 16. (a) Demonstration of actuation and self-deployable contracting cord metamaterial inspired by push puppets (reproduced from Ref. [161] with permission from the Royal Society of Chemistry.); (b_1) Applications of 3D metamaterial as novel smart trapper: the actuators offered perturbation to the metamaterial and made it retract rapidly in an omnidirectional mode after the target object is sensed, (b_2) Representation of shape-reconfiguration behavior of the 3D metamaterial including dynamic transformation, automatically achieved retracted state after being deployed in the initial state (process occurs very fast within 350.0 ms) (adapted from Ref. [163] copyright 2023 Wiley-VCH GmbH); (c) Representation of deployable, shape-morphing architectures conceptual applications in multipurpose reconfigurable space robots and habitats (adapted from Ref. [164] under a Creative Commons Attribution 4.0 International License); (d) Flat thick-panel Miura-origami 3D-printed sheets assembly for achieving self-locking property (adapted from Ref. [165] under a Creative Commons Attribution 4.0 International License).

7. Summary and prospects

In this review, we illuminate the emerging field that combines 4D printing with mechanical metamaterials, focusing on the innovative concept of shape morphing and energy absorption. We critically explore the multifarious properties of 4D-printed mechanical metamaterials and their potential to improve and innovate multifunctional applications across various engineering domains. Mechanical metamaterials research spans multiple disciplines, including mechanics, materials, mathematics, chemistry, biology, and electronics, thus facilitating multidisciplinary collaborations. Despite the advancements, current studies predominantly remain theoretical and lack practical applications. The following sections will outline key future areas for development in this promising field, as highlighted in Fig. 17.

- AM-based and design-based are the two primary challenges in the 4D printing of smart materials. The design-based challenges involve understanding the complexity of smart structures, especially in

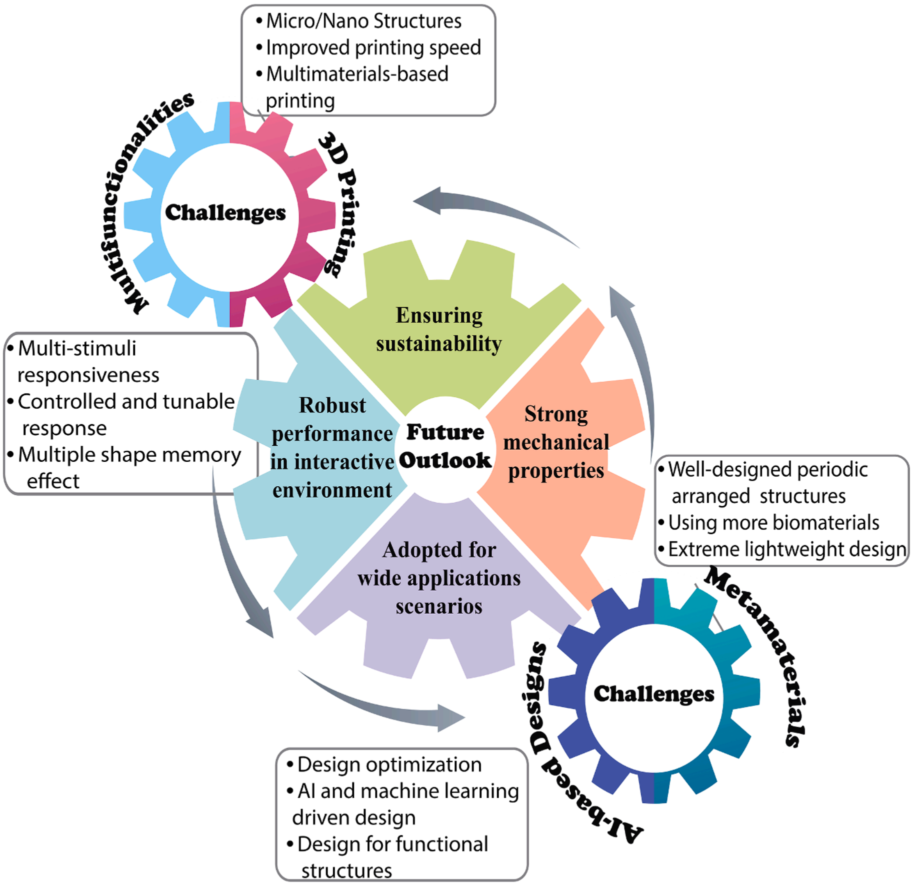

Fig. 17. Current challenges regarding printing techniques include 4D multifunctionalities and metamaterials enriched with artificial design for unlocking the true potential of 4D mechanical metamaterials.

engineered tissues, and addressing the limitation in achieving complex and controlled deformations, which impact the accuracy of smart products. These issues can be mitigated through computational approaches that predict the shape-morphing behavior of smart structures [55]. In the future, data-driven approaches will likely become the main framework, focusing on optimizing materials, structures, process control and tool paths. Thus, the integration of ML and AI is becoming a mainstream research area in mechanical metamaterials, aimed at ensuring highly optimized structures and more realistic designs. This interesting synergy of ML with 3D printing offers greater degrees of freedom in designing complex mechanical metamaterials for a variety of applications. Advanced ML technologies can further enhance print quality, reduce printing costs, and eliminate the need for complex constitutive modelling and computational approaches. This advancement leads to highly multiscale topology-optimized structures with greater design efficiency [167]. New domains within ML, such as deep learning, can mimic human cognitive processes and automatically improve learning by deriving rules from data without explicit programming, while others require over a million data points to train the model effectively [168]. Consequently, ML models are transforming the approach to rational materials design. These computational approaches play a promising role in designing novel art-inspired metamaterials, such as origami and kirigami, by incorporating advanced algorithms that combine folds and cuts [169].

- To address the challenges associated with 4D-printed metamaterials in biomedical applications, several safety concerns must be considered, including biocompatibility, biosafety (e.g., cytotoxicity studies on human cells) and mechanical stability, given that these are intended for long-term implantations. More information and clinical trials are necessary before these 4D-printed metamaterials can be deemed fully adaptable for advanced biomedical applications such as regenerative medicine, scaffolds, therapeutic devices, and implants that interact with and adapt to human biological conditions [170]. ML has contributed by analyzing large datasets related to the properties of various SMPs, cells, tissues, biodegradability, mechanical strength, and growth factors. AI provides insights into the outcomes of printing parameters and optimal biological conditions for specific tissue types. Additionally, integrating ML with AI can predict long-term cell behavior and tissue development, allowing researchers to forecast bioprinting outcomes and adjust parameters as needed [171]. Given AI's promising future, it is recommended to explore new ML algorithms to comprehensively investigate material-process-property relationships.
- It is worth mentioning that SMP 4D printed structures due to their inferior mechanical properties, constrained application environments, insufficient actuation force hinder shape recovery under loads, low energy output, and limited design freedom often missing commercial applications [172]. Moreover, commercial manufacturing of metamaterials-based smart devices meeting the standards of existing electronic devices is considered a grand challenge. Thus, the long-term durability of smart devices as well as economical and integrated mass production technologies need extensive attention for the complete translation of the 4D printed metamaterials-based electronic technologies into real-world applications. Moreover, the development of multi-stimuli-responsive-based reconfigurable mechanical metamaterials with dynamic bonding such as covalent and non-covalent dynamic bonds is quite challenging. The introduction of such dynamic bonds offers multifunctionality in terms of mechanical properties with self-healing

ability (the self-repairing and regeneration ability under external stimuli) and reprogrammable properties which broadens their applications in intelligent robots, wearable devices, human–computer interaction, and health monitoring [173].

- 4D-printed mechanical metamaterials exhibit remarkable properties such as stiffness, strength, shape morphing abilities, NPRs and extraordinary energy absorption properties due to the repeated arrangement of unit cells and size effects [174]. These properties are setting new standards for structural applications. Moreover, leveraging design freedoms in 4D printing by incorporating ML-based algorithms for structural optimization can enhance the density, dimensional accuracy, and shape-morphing ability of these metamaterials.
- Over the past few years, mechanical metamaterials have experienced significant advancements in designing sophisticated structures, such as foldable and self-deployable structures across multiple size scales and engineering fields. While moving towards the future, 4D-printed mechanical metamaterials are expected to revolutionize numerous engineering fields, from healthcare and consumer products to aerospace and defence. It is envisioned that these metamaterials will contribute towards more advanced systems with robust multi-functional performance and adaptability to complex environmental conditions [175].
- A major challenge for 4D-printed mechanical metamaterials is evaluating their shape-morphing abilities under multiple stimuli, as most current metamaterials are effectively under only one type of stimulus, such as heat. Another key challenge is accurately predicting structural deformation under various stimuli. Achieving controlled shape memory performance will enable precise control over actuation measuring parameters such as position, and curvature, which is crucial for designing smart actuators for practical applications. Moreover, smart materials employed for metamaterials often lack full control over deformation patterns, particularly at low scales, and frequently fail to replicate the desired shape-morphing behaviour, which is essential for developing self-deployable structures.

4D-printed mechanical metamaterials have now intersected with nearly all fields of science and technology, potentially leading to a more advanced and intelligent production mode. In conclusion, 4D-printed metamaterials represent a highly promising and forward-looking area of research, poised to shape the future of technological innovation.

CRediT authorship contribution statement

Muhammad Yasir Khalid: Writing – review & editing, Writing – original draft, Validation, Supervision, Software, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Zia Ullah Arif:** Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Formal analysis. **Ali Tariq:** Writing – review & editing, Visualization, Validation, Methodology, Investigation, Formal analysis. **Mokarram Hossain:** Writing – review & editing, Methodology, Investigation. **Rehan Umer:** Investigation, Methodology, Writing – review & editing, Funding acquisition. **Mahdi Bodaghi:** Writing – review & editing, Writing – original draft, Validation, Supervision, Project administration, Methodology, Investigation, Formal analysis, Conceptualization, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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