Research Article

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Quick insight into the dynamic dimensions of 4D printing in polymeric composite mechanics

Abstract: 4D printing is recognised for its numerous potential applications due to its reaction towards stimulus factors. However, limited research has focused on what, why, and how this stimulus-response works. This study reveals the mechanism used to stimulate 4D printing reactions. Complex printing via design structure and mechanical control on fibre orientations are promising techniques compared with chemical modifications, which are difficult to control, particularly for commercialisation.

Keywords: polymeric composites, shape-memory materials, 4D printing

1 Introduction

The fabrication of 4D-printed polymeric materials is frequently related to shape-memory polymers (SMPs), and it has been extensively discussed, particularly in the biomedical [1], construction [2], electronic device [3], food industry [4], and other fields [5]. SMPs are polymeric smart materials that can return from a deformed state to their original shape when induced by an external stimulus. These materials exhibit the ability to change size, shape, stiffness, or strain in response to various stimuli [6]. Meanwhile, the 4D printing of polymeric materials is a significant innovation in the field of additive manufacturing. It functions similarly to a conventional manufacturing process, i.e. injection moulding, thermoforming, and compression moulding. In general, SMPs introduce the fourth dimension, namely, time, allowing for the creation of dynamic and responsive structures that can adapt to changes in their environment over time. This technology is important due to its potential applications in various fields, i.e. robotics, biomedicine, and electronic devices. By using smart materials, such as SMPs, liquid crystal elastomers, composite hydrogels, and other responsive polymers, 4D printing enables the fabrication of complex, stimuli-responsive 3D structures, playing significant roles in the field of manufacturing. Water, thermal (heat), photo (light), magnetic field, chemical reaction [7], and pH are the major stimulus factors for 4D printing [1,8]. Stimulus factors play a role in shaping and transforming 4D-printed polymeric composites. Specific stimuli are required for shape and property changes in 4D-printed SMP composites. The choice of stimuli-responsive materials is crucial for determining the necessary stimuli for altering property and functionality in 4D printing. Researchers have investigated 4D printing as an emerging technology, emphasising the incorporation of the fourth dimension into conventional 3D printing. This integration enables printed components to switch between configurations triggered by external stimuli [9]. In addition, these stimulus factors are subjected to the shape memory effects experienced by printed polymers. In 2015, biomedical researchers collaborated with Stratasys in developing 4D printable SMPs subjected to time-dependent shape changes [10]. Stratasys utilises SMPs as a technology platform for biomedical applications, employing 4D printing to create items, such as cell scaffolds, vascular stents, bone scaffolds, tracheal stents and cardiac stents. The technology...
involves printing materials with varying water-absorbing capabilities, enabling the printed object to change shape when exposed to water. Essentially, various techniques, including deployable mechanisms, bi-stable structures, compliant mechanisms, assembly/disassembly, stimulus-induced deformation mismatch and shape-memory effect technology, enable 4D printing [11,12]. Affirmative steps through the years have refined 4D printing parts and the reactions of 4D printing towards stimulus factors [13]. On the basis of this understanding, factors that influence the 4D printing process include the type of additive manufacturing process, the type of responsive material, the type of stimulus, the interaction mechanism between the stimulus and the material, and the mathematical modelling of the material transformation. However, the reversibility of response in some polymers can impair the printability and reproducibility of the desired 4D effect in the product. Temperature-responsive polymers are amongst the most frequently used materials in 4D printing applications, particularly in the tissue engineering field, where a change in temperature can be easily controlled and applied in a non-invasive manner [14].

The primary inquiry revolves around the utilisation of SMPs in the fabrication of 4D-printed materials, primarily through their stimulus response in 4D printing. This stimulus-response plays a crucial role in determining the behaviour and functionality of printed polymer composite structures. In 2016, Ge et al. introduced a novel approach that facilitates high-resolution multi-material 4D printing by utilising highly customisable SMPs on a projection micro-stereolithography system. Their study also delved into the design of constituents and compositions to exhibit desired thermomechanical behaviour, enabling controlled shape memory behaviour [15]. This resource proves relevant for comprehending the integration of SMPs in 4D printing. Gonzalez et al. [16] found that determining materials for 4D printing is crucial because materials require self-assembly, or even multiple or designed materials. Fu et al. [8] agreed that the development of multi-materials provides benefits in managing dispersion and ability to control the response speed whilst meeting the deformation accuracy of commercial requirements. This capability enables improved adaptability through the integration of various materials with distinct properties and behaviour. Enhanced deformation accuracy provides more precise control over the deformation of printed components, resulting in improved response speed [17]. Given that 4D printing strongly depends on 3D printing, then understanding its major concerns, particularly non-symmetrical changing defects [2,18], poor mechanical performance [19], limited geometrical printer parameter [20], and limited materials and printer use has increased the difficulty of translating 4D printing into a production technology. However, some major concerns related to non-symmetrical changing defects and mechanical performance have also been raised. The use of multiple materials allows for more precise control over the deformation of the printed components, because each material can be designed to exhibit specific deformation behaviour under the influence of different stimuli. However, challenges exist, and they include material limitations, non-symmetrical changing defects, scaling up of manufacturing process, predictive models for 4D printing, and regulatory considerations [21,22]. Despite these challenges, the 4D printing of polymeric materials can exert a significant effect on the mechanical performance of composite structures. The use of smart materials, such as SMPs, liquid crystal elastomers, and composite hydrogels in 4D printing, enables the fabrication of complex, stimuli-responsive 3D structures that can adapt to changes in their environment over time. The mechanical properties of 4D-printed composite structures can be improved by introducing fibre reinforcement, which can significantly increase tensile strength.

To date, several studies have discussed applicable materials in 4D printing [23,24]; however, these studies are mostly related to shape-memory effects. The research gap reflects the importance of understanding the mechanism of 4D printing and its effects on mechanical performance [19,25].

2 Mechanism for inducing 4D printing of polymer composites

2.1 Cross-linking

In 2020, chemical cross-linking [1,8,26] and modification [25,27] have been required for establishing shape-memory effects because the deposited 4D print layers cannot be improved only through surface properties. These requirements clearly indicate that stimulus factors do not affect the motion of 4D-printed polymeric composites by themselves. Chemical cross-linking enhances the shape memory of thermoset SMPs. However, this improvement comes with a trade-off, i.e. reduced re-workability, because the permanent shape becomes fixed. The extent of cross-linking can affect the mechanical properties and shape memory behaviour of 4D-printed polymers. For example, in a study of 4D-printed continuous fibre-reinforced thermo-setting SMP composites, a curing agent content of 5% led to a moderate cross-linking degree, resulting in superior overall mechanical and shape-memory properties [28]. Two mechanisms are employed to implement cross-linking within
polymers: physical and chemical methods. Amongst chemical methods, radical polymerisation and irradiation-induced processes are the most common strategies. Gelation occurs as a result of the generation of reactive species through ionisation, typically induced by ultraviolet (UV) or electron beam irradiation. This leads to copolymerisation, where reactive functional groups within the formulation are utilised, forming a cross-linked structure in the printed samples due to covalent reactions within the functional groups of polymers, primarily hydroxyl, carboxyl, and amino groups [29].

In 2022, Chan et al. [27] used a photo-polymerisation technique on film samples in a UV box before printing the elastic structure by using digital light projection (DLP). The resin was added as a photo-absorber to minimise elasticity-induced position shifts whilst printing. Contrasting techniques and multi-materials are used on isolated cellulose fibres and graphene nanoplatelets (GnPs), in which cellulose fibre samples are initially 3D printed using direct ink writing (DIW) before controlling their hydrophilicity (water absorption from 94 to 81.2%) for 4D printing applications (Figure 1b) [30]. DIW involves layer-by-layer printing through extrusion, in which the nozzle moves to deposit viscoelastic ink. Additional techniques employed in DIW include frontal polymerisation, which initiates the polymerisation reaction, and reactive extrusion, which integrates chemical reactions into the extrusion process [31]. Meanwhile, a study that used carbon micro-fibre (CMF) hydrogel composite filaments suggested that manipulating fibre distribution and orientation by using nozzle actuation can enable motion stimulation [32,33]. The arrangement of fibres in 4D-printed composites influences

![Figure 1](image_url)

**Figure 1:** (a) Time-lapse images of swelling water-stimulus over 60 min. The blue text indicates the initial shape [32]. (b) Actual images of dehydration/hydration trigger stimulus induced by graphene content [30]. (c) Images of the printed PLA: (1) as-printed, (2) deformation at 70°, (3)–(5) recovery process within 20 s interval time, and (6) recovery-printed PLA [41]. (d) Comparison of experimental fused deposited modelling (FDM) and FEM under different deformation loadings for the (d-1) arc shape and (d-2) crescent shape, and (d-3) SMP processing of arc-shaped printed samples [42].
their mechanical characteristics and shape-changing behaviour. Researchers have devised methods for accurately modelling 3D-printed continuous yarns of fibre composites at the filament scale [34]. 4D-printed structures primarily rely on internal and residual stresses to induce reversible and one-time shape alterations. Furthermore, a design approach for the trajectory of fibres in the 4D printing of composite structures has been suggested to regulate deformation. The off-centre distribution of fibres in the composite can induce bending deformation under temperature stimulation. These advancements underscore the significance of fibre distribution in attaining the desired mechanical properties and shape-changing capabilities in 4D-printed composites [35]. Figure 1(a-1) and (a-2) clearly indicates that fibre distribution encourages motion stimulus (water mechanism) [32].

2.2 Magneto-responsiveness

The incorporation of magneto-responsive SMPs into 4D printing provides opportunities for creating innovative and intelligent products controlled through contactless methods. Yue et al. [36] reported that to stimulate magneto-responsive performance, cellulose nanofibres (CNF) were added to poly-hydroxybutyrate/poly(ε-caprolactone) (PBH/PCL). Their result revealed that the addition of CNF and their ability to control 3D-printing parameters indicated high magneto-responsive performance (21 s shape-recovery time) with 55.13% tensile strength increment compared with hot-pressed samples [36]. Studies have indicated that the role of incorporating CNFs into magneto-responsive SMP composites, along with Fe₃O₄, yields materials with outstanding mechanical properties [37,38]. When magnetic nanoparticles are integrated into the polymer matrix, the resulting cellulose materials exhibit interesting mechanical behaviour that is dependent on the magnetic field. Furthermore, CNFs integrated with magnetic nanoparticles have been employed to produce membranes with magnetised high toughness [38]. Previous studies have demonstrated the integration of magneto-responsive materials, including ferromagnetic droplets, magnetic shape memory composite hydrogels, magento-thermally deformable SMPs, and magnetic particle-driven composite elastomers, into 4D printing [39]. These materials enable the creation of innovative and mechanically robust structures with remote actuation capacity and shape-changing abilities, achieved through the application of magnetic fields. The filler consists of magnetic materials that respond to external magnetic fields, enabling the desired shape changes and functionalities. The mechanism for applying magneto-responsiveness has been elaborated in prior studies [40]. In this mechanism, the filler behaves similarly to that in magnetometry, with billions of nanoparticles densely packed and magnetically oriented at the core of the droplet, imparting magnetic properties to the entire droplet [40].

2.3 Thermal

In the past years, numerous strategies have been used to realise material processing and its effects on 4D printing. In 2022, Akbar et al. [18] discussed the importance of 3D printing parameters on shape-memory effects. Notably, other relevant mechanisms (Table 1) that contribute to stimulation behaviour are yet to be discussed because the literature (<3 years) has focused on stimulus factors. Figure 1(c-1)–(c-6) shows the thermal stimulus response induced by using fibre orientation during extrusion printing [41]. One study exhibited that the printed samples depended not only on temperature as the primary stimulus factor but also on the time required to change the recovery phase into the original shape. Notably, the deformation mode (bending and stretching) leads to stress deformation that mimics biomaterial applications. This phenomenon was discovered by Xin et al. [42] whilst using geometric parameters. Another study that used polylactic acid (PLA) materials considered the arc shape and crescent shape whilst controlling deformation and Poisson’s ratio (Figure 1d). The results indicated that the maximum strain was ~90% when considering torsion-compression during the temporary shape [43]. Moreover, the arc shape and crescent shape exhibited similar nonlinear deformation behaviour as potential application (i.e. lens capsule, foetal membrane, pig belly skin, and iliac artery) when programming and reconfiguring the deformation (λ) [42]. Compared with other polymer materials, PLA has been recognised for its ability to exhibit the shape memory effect within a specific temperature range. The hard segment in PLA is elastic, whilst the stiffness of the soft segment is appropriately balanced. By adjusting temperature, PLA can maintain a permanent shape in a glassy state below its glass transition temperature (T_g). Above its T_g, PLA becomes sufficiently pliable to undergo deformations into transient, alternative forms. The temporary shape achieved through deformation is preserved as long as the temperature remains below T_g. Furthermore, the initial shape can be stored, and PLA can be reheated above T_g to recover the original shape by removing applied forces [44–46]. Notably, amongst various polymer materials, PLA stands out as the most commonly utilised in 4D printing technology. This recognition is attributed to its ability to be synthesised, with 90% of PLA obtained from the bacterial fermentation of sugar, by employing
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<tr>
<td>Isolated cellulose fibres/GnPs</td>
<td>Hydrophilicity (water absorption)</td>
<td>– Water</td>
<td>DIW</td>
<td>– 80.28 ± 0.99 MPa (Tensile stress)</td>
<td>[30]</td>
</tr>
<tr>
<td>CMFs hydrogel</td>
<td>Nozzle actuation – orientation behaviour</td>
<td>– Water</td>
<td>FDM</td>
<td>– 60.67 MPa (Tensile strength)</td>
<td>[32]</td>
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<tr>
<td>PBH/PCL-reinforced CNFs</td>
<td>Composition (0.5 wt% CNF)</td>
<td>– Magnetic field</td>
<td>FDM</td>
<td>– 2.1 s (Shape recovery time)</td>
<td>[36]</td>
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<tr>
<td>Carboxymethyl cellulose (CMC)/clay/citric acid</td>
<td>Crosslinkable polymer</td>
<td>– Ultrasonication</td>
<td>FDM</td>
<td>– 1.15 kPa (Modulus of elasticity)</td>
<td>[48]</td>
</tr>
<tr>
<td>Continuous carbon fibres/polyactic (PLA)</td>
<td>High extruder temperature (200–230°C)</td>
<td>– Electric induced</td>
<td>FDM</td>
<td>– 75 s recovery time (5 V)</td>
<td>[49]</td>
</tr>
<tr>
<td>Polyurethane-based thermoplastic (grade MM-5510)</td>
<td>Glass transition ($T_g$)</td>
<td>– Thermal</td>
<td>FDM Simulation</td>
<td>– 40 MPa (Residual stresses)</td>
<td>[18]</td>
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<td>Shape memory fibre (types DM9850 and DM9885)</td>
<td>– $T_g$</td>
<td>– Thermal</td>
<td>FDM Simulation</td>
<td>– 0.6–0.7 mm (Measured warpage)</td>
<td>[2]</td>
</tr>
<tr>
<td>Poly(ethylene-glycol-adipate) diols/diphenylmethane disocyanate and diethylene glycol</td>
<td>Crosslinkable polymer</td>
<td>– Electrostatic induction</td>
<td>FDM</td>
<td>– 56 mW·m$^{-2}$ (Power density)</td>
<td>[50]</td>
</tr>
<tr>
<td>Continuous fibre-reinforced composites</td>
<td>– $T_g$</td>
<td>– Thermal</td>
<td>FDM</td>
<td>– 27.37 MPa (Tensile strength)</td>
<td>[20]</td>
</tr>
<tr>
<td>Thermoplastic polyurethane (TPU)/PLA</td>
<td>Microfiber content</td>
<td>– Thermal</td>
<td>Extrusion based</td>
<td>– 87.36 MPa (Breaking strength at 16.32 wt% fibre content)</td>
<td>[41]</td>
</tr>
<tr>
<td>Multi-stable metamaterials</td>
<td>– $T_g$</td>
<td>– Thermal</td>
<td>Light-curable inkjet</td>
<td>– 81.1–86.5% (shape recovery ratio, $R_t$ is between 1 and 7 s)</td>
<td>[51]</td>
</tr>
<tr>
<td>– Monomer benzyl methacrylate</td>
<td>Crosslinkable polymer</td>
<td>– Thermal</td>
<td>Laser based</td>
<td>– 24 MPa (Tensile strength)</td>
<td>[52]</td>
</tr>
<tr>
<td>– Multi-functional methacrylate poly (ethylene glycol) dimethacrylate</td>
<td>Printing parameter</td>
<td>– Thermal</td>
<td></td>
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</tbody>
</table>
common polymerisation methods, such as ring opening and condensation. Commercially recognised for its success in 3D printing technology, PLA has gained widespread usage, particularly in medical and electronic applications [44]. A study on tissue generation applications reported that 3D printing evidently managed to mimic osteochondral tissues, which were later strengthened with the use of fibres as reinforcement materials [47]. A similar mechanism has been used by a few researchers to stimulate the motion effects. The results of which have been discussed in Section 3.

3 Mechanical and electrical performance of 4D-printed polymer composites

After determining the ability of stimulation factors that affect the mechanical performance of printed polymeric composite samples, a series of study has been conducted to advance the understanding of 4D-printed samples. Dong et al. [20] reported that by adjusting the structural design of printed samples, PLA composites with a maximum of 27.3 MPa were recorded. During the 4D stimulus response, evidence of buckling deformation existed without an increase in stress, because stress plateaued from 0.005 to 0.01 mm·mm\(^{-1}\), indicating that the study was successful. Moreover, the study reported that tensile performance was subjected to printed layer thickness. As layer thickness increased, tensile strength started to deteriorate (maximum of 90 MPa at 0.1 mm), because more printed layers would enforce larger fibre content at lower height, strengthening the structure and resulting in higher strength. On the basis of this understanding, the mechanical properties of 4D-printed polymeric composites are significantly influenced by printing temperature and layer thickness. Research indicates that increasing printing temperature can improve the mechanical properties of materials, such as PLA and polyethylene terephthalate glycol (PET\(_g\)).

### Table 1: Continued

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<tr>
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<tr>
<td>– Phenylbis (2,4,6-trimethyl benzoyl) phosphine oxide</td>
<td>– Lattice structure</td>
<td>– Thermal</td>
<td>FDM</td>
<td>– 100% (maximum strain of lattice structure)</td>
<td>[53]</td>
</tr>
<tr>
<td>PLA</td>
<td>– Angle-ply (design structure)</td>
<td>– Thermal</td>
<td>FDM</td>
<td>– Average recovery rate 1.1·s(^{-1}) (for 90° printed sample)</td>
<td>[54]</td>
</tr>
<tr>
<td>PLA</td>
<td>– Crosslinkable polymer</td>
<td>– Water</td>
<td>DLP</td>
<td>– 2–15 MPa (tensile strength from 55 to 80°C)</td>
<td>[55]</td>
</tr>
<tr>
<td>TPU</td>
<td>– Material (thermochromic pigment powder)</td>
<td>– Light transmission spectra</td>
<td>DLP</td>
<td>– 96.4% (shape fixity)</td>
<td>[56]</td>
</tr>
<tr>
<td>Resin (DentaClear)</td>
<td>– Water content</td>
<td>– UV-C irradiation</td>
<td>Extrusion based</td>
<td>– 10.52–29.53% of reduction in average light transmission intensity (3D printing)</td>
<td></td>
</tr>
<tr>
<td>Ergosterol</td>
<td>– Water content</td>
<td>– UV-C irradiation</td>
<td>Extrusion based</td>
<td>– 6.51% of reduction in average light transmission intensity (4D printing)</td>
<td></td>
</tr>
<tr>
<td>– 1.2 μg·g(^{-1}) (vitamin D(_2) concentration) and 3.5 mg of ergosterol consumption at 4 h exposure time</td>
<td></td>
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<td></td>
<td>[4]</td>
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</tbody>
</table>
because it reduces the viscosity of the melts and strengthens the fusion between layers [57]. In addition, layer thickness affects the mechanical properties of polymers, with some studies indicating that increasing layer thickness can increase the tensile strength of a material [58,59]. These findings underscore the importance of carefully controlling printing temperature and layer thickness to achieve the desired mechanical properties in 4D-printed polymeric composites.

For thermal stimulus response, a study on thermomechanical analysis was performed to understand the mechanical performance of polymeric materials during 4D printing process. This study reported that the spin crossover polymer was able to sustain up to 220°C without property interference [60]. However, a stiff drop (~75%) was recorded between Young's modulus at room temperature (1.7 ± 0.2 GPa) and 0.2 GPa at 90°C [60]. This finding indicated that the damping behaviour was experienced by the printed polymer, because the discontinuity curve was visible until 80°C. This result is attributed to the viscoelastic motion of extended defects under dislocations, stretching under tension whilst minimising diffusion during compression [61]. One method of inducing thermal stimuli involves the incorporation of carbon fibres given their high thermal conductivity, enhancing heat transfer within composite materials. In a study conducted on continuous pitch carbon fibre composites, a maximum effective thermal conductivity of 37.1 W·m⁻¹·K⁻¹ was observed for a 9.5% volume fraction of pitch carbon fibre [62]. In addition, the inclusion of carbon fibres can affect the mechanical behaviour of 4D-printed materials at elevated temperatures. Research has indicated that the interfaces and performance of printed composites are influenced by process parameters, such as temperature and pressure. The study further revealed that the transverse thermal conductivity of carbon fibres increased with a decrease in the volume fraction of the fibres, whilst in-plane thermal conductivity increased with an increase in volume [63]. A study on magnetorheological elastomer (carbon-deposited PLA) printed with Ultimaker S3 FDM-type 3D printer in the form of concentric filling indicated that the $T_g$ recorded reached as close as 75°C compared with pure PLA (55°C) (Figure 2a) [64]. This finding suggested that when considering 4D printing, one should know additive addition will modify $T_g$ due to the heterogeneous nucleation locations that raise PLA crystallisation temperature. Figure 2(c) displays the reported data obtained when incorporating magnetic fillers into polymeric composites, particularly PLA and TPU. A slight decrease was observed as the magnetic filler ($Fe_2O_4$) was added, reaching a minimum of 48 MPa for tensile strength at 30 wt%. Meanwhile, elastic modulus increased from 1,291 to 1,452 MPa [39]. This phenomenon is attributed to the increased crystallinity of PLA and inorganic particles. Moreover, carbon addition induced greater stiffness of the PLA matrix, resulting in higher storage modulus (2,500 MPa). In conjunction with these findings, the voltage applied was varied when subjected to $T_g$. This phenomenon is due to the fact, that if the voltage increase beyond 60 V, then temperature will be uncreased by up to 90°C (at 120 V). As the samples heat up (120 V) during the process, resistance will increase (8,500 Ω), deteriorating electrical performance. The manipulation of voltage levels in 4D-printed carbon-deposited PLA composites can give rise to shape-memory effects, allowing for controlled shape-recovery behaviour when subjected to an applied direct current voltage. Thermal images taken after applying a constant voltage to printed samples revealed the electro-induced shape memory effect, suggesting the feasibility of utilising electrical stimuli to govern the shape memory characteristics of these composites. Moreover, the introduction of continuous carbon fibres augments the bending resistance of the composites, emphasising the possibility of customising mechanical and shape-memory properties by adjusting voltage and incorporating carbon fibres [46]. The results demonstrated that the structure returned to its initial shape as soon as temperature passed $T_g$ (17 s of total time) [64]. Lalegani Dezaki et al. [65] extended their research by using SMP materials with a dynamic tanδ ranging from 0.4 to 0.6. The stress–strain analysis recorded super elastic behaviour up to 30% strain, because pores decreased as load was applied. This result indicates that having a foam structure will allow greater stiffness because foam is compacted by the process. Utilising foam structures in 4D-printed polymers provides benefits, such as lightweight design, enhanced stiffness-to-weight ratio, customised mechanical properties, efficient energy absorption, damping capabilities, and adaptive responses. Research has indicated that 4D-printed SMPs that feature a wide range of glass transition temperatures can be programmed to adopt multiple temporary shapes, facilitating intricate shape alterations in response to various stimuli. In addition, the temperature-dependent mechanical behaviour of 4D-printed composite lattice structures, reinforced by continuous fibres, exhibits the adaptive transformation of morphology, structure, and behaviour when exposed to external stimuli. In addition to the experimental study, analyses that used the finite element method (FEM) were conducted for glassy polymer (VeroBlack) and elastomer (TangoPlus) by considering deformation occurrence [66]. The findings indicated that the bending angle recorded (via shape-recovery ratio) exhibits an excellent correlation with FEM simulation (ABAQUS). Notably, the increment in thickness caused lesser bending angles but minimum errors were recorded when compared with FEM analysis (maximum error of 9.8%) [66]. On the basis of this study, detailed simulation analysis determined that high-resolution complex 4D
printing resolves the inverse issues whilst embodying a wide range of applications [66,67]. In general, the bending angle of 4D-printed polymers tends to decrease after numerous cycles of shape recovery, primarily due to material fatigue and degradation. The repetitive deformation and recovery process can cause damage to the internal structure of the polymer, resulting in diminished mechanical performance and a decline in bending angle. Notably, the degree of change in bending angle is contingent upon the particular material and design of the printed structure [66]. In 2023, Lalegani Dezaki and Bodaghi [68] reported that the unloading structure was unable to fully return to the original shape. This
result strongly recommends conducting a future study on bending angle upon shape recovery after a certain cycle. One study suggested that when heat (70°C) and negative pressure (vacuum) were applied, the meta structure actuator (Figure 2b) experienced a stiff structure (between 49 and 74 N in accordance with the cyclic load graph). Compared with a non-vacuumed and paired actuator, the stiff structure was maintained given that no energy was absorbed by the structure because it is in a rubbery stage [69]. The utilisation of a vacuum in combination with the shape-memory effect empowers polyurethane meta-structure actuators to function as soft/hard robots through hot and cold programming, complemented by negative air pressure. When subjected to vacuum pressure, substantial normal forces arise between internal components, generating frictional forces that prevent the jamming structure from deforming. Actively managing pressure difference between the inside and outside of the vacuum bag enables the control of the structure’s rigidity, enhancing stiffness. This method offers the benefit of not requiring continuous negative air pressure for actuation, contributing to the sustainability and efficiency of the actuation process [70]. Thus, an extensive study is required, particularly to correlate the mechanism used to induce 4D printing and its effects towards the mechanism. The structure of the printed sample significantly influences its mechanical performance during 4D stimulus response. Studies have shown that the addition of functional fillers or the construction of bilayer structures by using different materials with varying response properties can enhance the responsiveness and shape-shifting capabilities of 4D-printed structures [71].

4 Future prospect

The translation of 4D printing into production technology encounters notable challenges, including material limitations, intricate fabrication processes, and regulatory considerations. The complexity of fabrication involves the amalgamation of diverse materials, achieving precision, and ensuring scalability and automation for large-scale industrial production. Regulatory aspects entail the establishment of standards to ensure the safety and efficacy of 4D-printed products. Overcoming these challenges is pivotal for the successful transition of 4D printing from research laboratories to industrial production [21,72]. Future research in 4D printing mechanisms and their effects on polymeric composites can explore innovative actuation methods, such as reversible 4D printing actuation by using elastomer swelling and heat, as proposed in a recent study [73]. In addition, the taxonomy of shape-changing behaviour for 4D-printed components, which categorises shape change behaviour into basic shape change, complex shape change, and a combination of shape changes, provides a framework for further research into understanding and leveraging the diverse shape-changing capabilities of 4D printing. Furthermore, the integration of vacuum-based jamming with the shape memory effect of 4D-printed metamaterials to reduce energy consumption and enhance actuation behaviour presents a promising avenue for future research in developing sustainable and efficient soft/hard robots through hot and cold programming accompanied by negative air pressure. These research directions aim to advance the understanding and utilisation of 4D printing mechanisms and effects on polymeric composites by exploring new actuation methods, classifying shape-change behaviours, and enhancing actuation behaviour through the integration of vacuum-based jamming and shape memory effects. This research exhibits the potential to drive innovations in the development of smart composites, advanced actuation techniques, and sustainable soft/hard robotic systems.

5 Conclusion

This study discusses the mechanism for inducing 4D stimulus responses, particularly by using polymeric composite materials. To date, no mechanism is simpler than altering the cross-linked polymer or understanding the glass transition temperature of materials. Replicating similar processes for commercialisation purposes is clearly difficult. Nozzle actuation that promotes fibre orientation or even design structure during printing appears to be the best option for aiding the stimulus responses of printed materials. Further study is necessary to strengthen the mechanism, particularly via mechanical tools or aided simulation design, because at present, only one paper has discussed this topic by using nozzle actuation. Future studies will enhance and allow the industry to control output performance.

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