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4D printing roadmap

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4D Printing Roadmap

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Abstract

Four-dimensional (4D) printing is an advanced manufacturing technology that has rapidly emerged as a transformative tool with the capacity to reshape various research domains and industries. Distinguished by its integration of time as a dimension, 4D printing allows objects to dynamically respond to external stimuli, setting it apart from conventional 3D printing. This roadmap has been devised, by contributions of 44 active researchers in this field from 32 affiliations world-wide, to navigate the swiftly evolving landscape of 4D printing, consolidating recent advancements and making them accessible to experts across diverse fields, ranging from biomedicine to aerospace, textiles to electronics. The roadmap's goal is to empower both experts and enthusiasts, facilitating the exploitation of 4D printing's transformative potential to create intelligent, adaptive objects that are not only feasible but readily attainable. By addressing current and future challenges and proposing advancements in science and technology, it sets the stage for revolutionary progress in numerous industries, positioning 4D printing as a transformative tool for the future.

Keywords: 4D Printing; Additive Manufacturing; Smart Materials; Active Materials; Adaptive Structures.

1 - Emergence of 4D Printing

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In recent years, four-dimensional (4D) printing has emerged as an innovative technology that provides unprecedented opportunities across a wide variety of research domains while gradually linking to industrial sectors. This paradigm shift from conventional three-dimensional (3D) printing lies in its ability to integrate time as a dimension, resulting in objects that dynamically respond to external stimuli. In the context of 4D printing, the term 'fourth dimension' pertains to the time-dependent response of printed objects to external stimuli. This sets it apart from typical three-dimensional printing, which primarily focuses on the process of additive manufacturing. It is crucial to comprehend that 4D printing utilizes 3D printing technology to physically produce objects, but it also adds a fourth dimension that allows objects to change or adapt over time in response to environmental changes, thereby introducing dynamic functionality. Moreover, the processing parameters of 3D printing, such as the speed of production or the temperature of the settings, have a significant impact on the behavior of these dynamic structures once they are made. The dependency of 4D printing capabilities on the foundational 3D printing parameters highlights the relationship between the two. This explains why adjustments to the manufacturing process can have a major impact on the functional outcome of 4D applications.

The pace at which 4D printing is advancing is nothing short of remarkable, propelled by innovative materials and their diverse applications. The timeliness of this roadmap could not be more crucial, as it aims to consolidate the rapid developments in 4D printing, making them accessible to experts in various fields, from biomedicine to automotive, textiles to electronics. Our roadmap serves as a vital compass for navigating this burgeoning landscape, offering insights into the latest breakthroughs, from the use of shape memory polymers, metamaterials, and liquid metals to the moldless manufacturing of composites and the integration of piezoelectric materials. It underscores the far-reaching impact of 4D printing, fostering efficiency, adaptability, and innovation in industries that affect our everyday lives. This roadmap is poised to empower experts and enthusiasts alike to harness the transformative potential of 4D printing, steering us toward a future where smart, dynamic objects are not only possible but readily accessible. It is structured to encompass the vast and evolving field of 4D printing technology, systematically covering key topics to offer an in-depth look. The structure of the roadmap is as follows:

Advancing 4D Printing with Innovative Materials and Applications

Sections 2–8 form a cohesive cohort that dives into the material science aspects of 4D printing. These sections discuss various materials used in 4D printing, including shape-memory metamaterials, liquid metal, NiTi shape memory alloys, composites, functionally graded additive manufacturing for shape-changing textiles, and piezoelectrics. These sections emphasize the role of innovative materials in enhancing the resilience and sustainability of products through 4D printing techniques.

4D printing, at its core, hinges on a programmable structure enabled by additive manufacturing (AM) techniques and functional materials. Liquid metals [1], with their unique properties, are emerging as materials of interest for 4D printing, enabling dynamic conductive components and circuits, potentially revolutionizing electronics. Composite 4D printing [2] introduces moldless manufacturing techniques, curving structural components made from high-strength materials, offering a cost-effective approach for diverse industries. Furthermore, the integration of AM and textiles is fostering 4D textiles [3], where elastic fabrics and printed structures synergize to create shape-changing textiles. The incorporation of piezoelectric [4] and thermoplastic [5] materials in 4D printing paves the way for precise shape changes in response to electrical stimuli, revolutionizing fields like soft robotics and morphing structures. In the 4D printing landscape, innovative materials and applications are driving transformative advances, underscoring the technology's profound potential in various industries.

The Transformative Potential of 4D Printing in Robotics, Sensors, Actuators, and AI Control

Sections 9–13 explore specific applications of 4D printing in the fields of robotics, sensors, actuators, and control systems. This section highlights the integration of 4D printing technologies into soft robotics, the development of dielectric elastomer actuators, and bio-inspired musculoskeletal systems, showcasing the practical and functional applications of 4D printing. It also discusses AI-based control in 4D printing, highlighting the synergy between artificial intelligence and 4D printing for smarter and more adaptive manufacturing processes.

The advent of 4D printing technology has introduced novel design approaches and AM techniques, enabling new age in the field of robotics. Soft robotics, a specialized branch of robotics, centers on the development of robotic systems constructed from flexible and deformable materials. The fusion of soft robotics with 4D printing has paved the way for substantial advancements. Notably, it has harnessed smart materials and computational modeling, fostering the creation of 4D-printed soft robots and actuators with intricate shapes and dynamic capabilities [6]. Leveraging machine learning (ML) techniques has enabled the anticipation of actuation mechanisms, propelling this field towards precision and autonomy. Additionally, the integration of artificial intelligence (AI)-based control strategies has started to address the challenges posed by the nonlinear nature of soft robotics [7]. Concurrently, 4D printing has revolutionized the domain of dielectric elastomer actuators (DEAs) [8], streamlining their fabrication, enhancing reliability, and expanding their multifunctionality. These actuators, responding to applied voltage, are integral components of smart structures, often coalescing with sensors and conductors, engendering versatile applications in soft robotics, human-machine interaction, medicine, vibroacoustics, and optics [9]. Furthermore, multimaterial 4D printing has unlocked the potential to engineer complex musculoskeletal systems [10], bridging the gap between rigid and soft robotics. This technology harmoniously combines active and passive materials, exemplified by shape memory polymers and conductive polymers, within a single framework, offering multifunctionality, sensory capabilities, and energy harvesting in a unified robotic system. The dynamic field of AI-based control in 4D printing [11] is optimizing the performance of these intricate robots, evolving towards autonomous decision-making. Within the domain of soft sensors [12], 4D printing has introduced efficiencies in crafting sensors with capabilities like piezoelectric, piezoresistive, capacitive, and triboelectric responses. These sensors fabricated using diverse AM techniques, open new avenues in wearable electronics and human-machine interfaces. Notably, the emergence of magnetoelectric tactile sensors through 4D printing showcases the capacity to convert mechanical pressure into electrical signals, aligning seamlessly with the principles of 4D printing. In this era of innovative materials and methodologies, 4D printing is poised to revolutionize soft robotics, sensors, actuators, and AI control, promising a future marked by adaptable and intelligent robotic systems.

4D Printing in Biomedicine and Health

Sections 15–17 are dedicated to the bio-medical applications of 4D printing, specifically focusing on 4D bioprinting and its role in health and pathophysiology. This segment covers the groundbreaking potential of 4D printing in cancer therapies and other medical applications, demonstrating the critical impact of 4D printing in revolutionizing medical treatments and diagnostics.

In the realm of biomedicine, tissue engineering, and cancer therapies, 4D printing has emerged as a transformative technology, offering unprecedented precision and control in fabricating intricate structures with dynamic functionality [13]. 4D bioprinting, a subset of 4D printing, signifies a significant leap in tissue engineering. This approach introduces the "fourth dimension" into the creation of constructs containing living cells. This dimension encapsulates the ability of these constructs to undergo deliberate transformations in shape or functionality in response to external stimuli. While still in its proof-of-concept phase, 4D bioprinting holds immense promise for applications ranging from organ transplantation to anatomical modeling and surgical implants. Cell traction force are harnessed to craft intricate 3D structures from 2D cell sheets responding to stimuli such as changes in ion concentration, temperature, magnetic fields, or near-infrared (NIR) light. Expanding on 4D printing's capabilities, 4D biofabrication aims to create structures using both living cells and non-vital materials. This novel approach leverages spontaneous transformations, including maturation and shape changes, within fabricated living objects. Synchronized transformations in multiple objects enable the creation of complex 3D shapes, offering unique possibilities for fabricating vascular networks and intricate patterns, such as those found in blood vessels atchara [14]. Cancer, a complex and deadly disease, has been historically treated through surgery, radiotherapy, and chemotherapy. However, the emergence of 4D printing technology is revolutionizing cancer therapeutics [15]. By offering personalization, controllable dosing, and site-specific treatment, 4D-printed structures, including scaffolds, devices, and robots, are addressing some of the most challenging aspects of cancer management. The field of pathophysiology, devoted to understanding and addressing disorders ranging from cardiovascular diseases to neuro-injuries, is increasingly turning to 4D printing for novel solutions [16]. By harnessing shape-changing materials and AM techniques, 4D-printed devices can mimic biological tissues, respond to specific stimuli, and trigger tailored biological responses. This convergence of innovation offers the potential to reshape the landscape of pathophysiology management, offering customized solutions for improved therapy outcomes. In light of these transformative possibilities across various facets of biomedicine, this roadmap embarks on a journey through the multifaceted world of 4D printing, exploring its current state of the art and future directions. Each section delves into a unique dimension of this evolving field, shedding light on the promise it holds for the betterment of human health and the advancement of medical science.

High-Resolution Micro- and Nano-4D Printing for Precise Smart Structures

Sections 18–19 address advanced technologies and design methodologies in 4D printing, ranging from micro- and nano-4D printing to high-resolution 4D printing.

While 4D printing has made significant strides at the macro scale, the burgeoning demand for precise micro- and nanostructures with adaptable properties in fields such as biomedicine, micro-optics, and micro-robotics has necessitated pioneering work in high-resolution [17] micro- and nano-4D printing [18]. Among various 3D printing methods, light-based techniques, particularly two-photon laser printing (2PLP) [18], have emerged as a frontrunner, enabling nanoscale manufacturing with unparalleled precision. This technology leverages femtosecond-pulsed high-energy lasers to induce spatially confined two-photon polymerization in photo-reactive materials, achieving resolutions well below the micron. Recent research has focused on the design of stimuli-responsive materials to unlock the full potential of micro- and nano-4D printing. These advancements mark a pioneering effort in the realm of high-resolution micro- and nano-4D printing, heralding the development of precise and intelligent microstructures with multifunctional capabilities.

Evolution of 4D Printing in Design and Artefact Development

Section 20-24 This segment also delves into accelerated design processes, pattern-controlled printing, topology optimization, and multi-axis 4D printing. These chapters highlight the cutting-edge technological advancements and sophisticated design techniques that are pushing the boundaries of what is possible in 4D printing.

The rapid evolution of 4D printing has been the result of collaborative efforts in materials science, mechanical modeling, and chemistry, culminating in the creation of multifunctional structures driven by active materials and energy stimuli. Researchers have advanced the deposition and performance of active materials, crucial for industry adoption. Yet, the integration of smart materials and AM into adaptable, shape-changing objects presents a substantial challenge, engaging both design and engineering disciplines. Recent endeavors have centered on multimaterial 4D printing, merging passive and active materials to enhance actuation and achieve intricate shape transformations [19]. This integration necessitates advancements in material deposition techniques and computational design tools. The emergence of voxel-based modeling has facilitated precise material distribution, enabling the prediction of shape changes in response to energy stimuli. Simultaneously, innovative techniques, combining genetic algorithms, machine learning, and topology optimization, have streamlined material distribution for complex shape changes. Pattern-controlled 4D printing [20] has emerged as a critical aspect, offering precise control over permanent or temporary shape changes through diverse pattern categories, from infill patterns to origami-inspired designs. Finally, 4D artifacts represent a transformative category, encompassing objects capable of self-deformation, healing, or assembly through external stimuli. Digital manufacturing technologies serve as the foundation for democratized 4D artifacts, with ongoing research aimed at customized materials, design tools, and post-processing techniques to expand possibilities and enhance interactivity. The integration of 4D technology throughout the artifact lifecycle promises disruptive changes in traditional manufacturing, introducing opportunities and challenges for sustainability.

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2 - Shape-Memory Metamaterials by 4D Printing for a Resilient and Sustainable future

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Status

The realm of 4D printing, particularly within the context of thermo-mechanical metamaterials boasting shape recovery and energy absorption/dissipation attributes, stands as a beacon of innovation, sustainability, and transformative resilience [1]. This section offers insights into the current state and the pivotal role it plays in fostering sustainable and net-zero practices while enabling diverse applications.

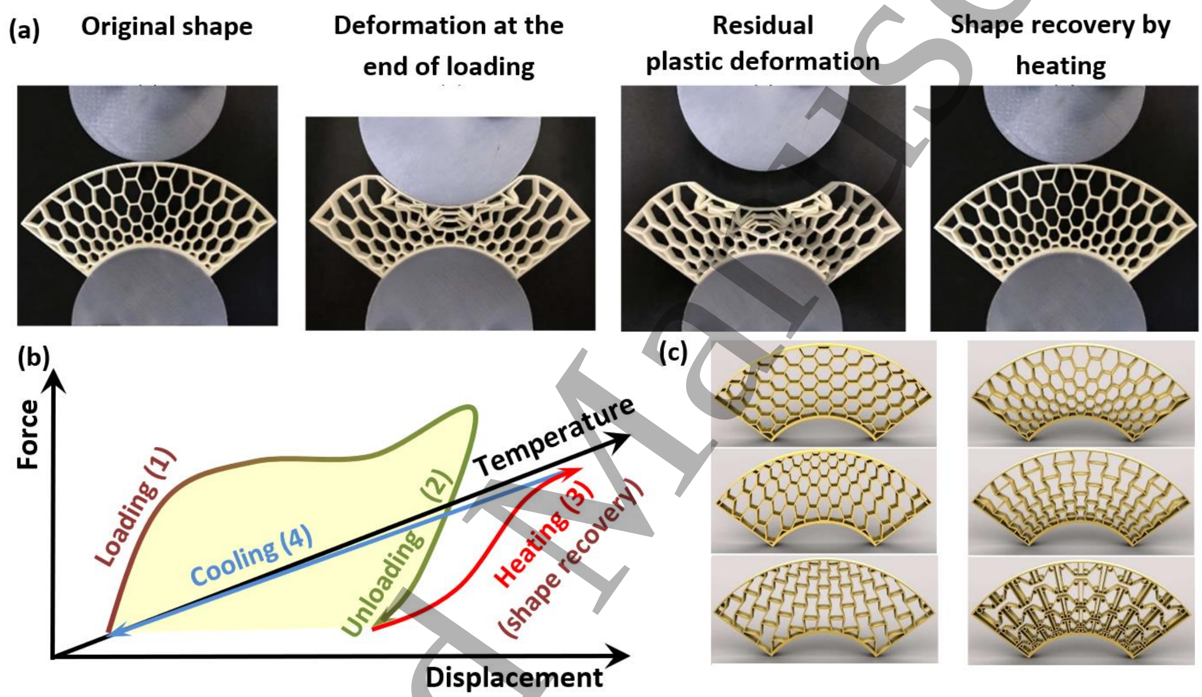

Figure 1. (a) Configuration of the 4D printed metamaterial before loading and at the end of loading, unloading, and heating-cooling; (b) a 3D force-displacement-temperature schematic diagram of the metamaterial's behaviors; (c) various metamaterial lattice patterns.

Since 2017, our pioneering research team with a vision of resilience and sustainability have initiated breakthroughs in developing lattice-based metamaterials with shape memory polymers (SMPs) through 3D/4D printing technologies for sustainable and eco-conscious practices, see e.g., [2-11]. The 4D printed metamaterials show remarkable energy absorption/dissipation, shape recovery, and adaptability, for instance as shown in Figure 1. The metamaterial is first 4D printed and then is mechanically loaded at a temperature below glass transition (T_g) and beyond the SMP yield stress causing strain softening and plastic flow (plateau) followed by hardening in both material and structural scales (step 1 in Figure 1b). Yielding and strain softening in the material scale happens when the stress overcomes the resistance of intermolecular segmental rotation, and further deformation stretches the crosslinked network/chains resulting in a strain hardening. Plasticity occurs and chains reorient along the loading direction while dissipate/absorb the energy. In the structural scale, the struts buckle by the mechanical loading, and the collapse softens the structure (strain softening, Figure 1a). By further loading the metamaterial flows into the collision location creating a denser structure with resistance and hardens the meta-structures. Upon unloading (step 2 in Figure 1b), the SMP at material and structural scales releases some of the absorbed energy and elastic deformation while a plastic residual deformation remains into the material/structure (Figure 1a and 1b). The area below force-displacement graph is considered as energy dissipation. The next step is to fully recover the original shape achieved by heating above T_g (step 3 in Figure 1b) followed by cooling to the initial temperature (step 4 in Figure 1b). The lattice topology for example as shown in Figure 1c can be tuned to control the mechanical behaviors of the metamaterials (e.g., force-displacement curve). The metamaterials' advantages are briefly listed below:

Shape Recovery and Reversibility: This feature [2, 3] makes them resilient and reusable, and ideal for applications demanding longevity and sustainability.

Efficient Energy and Material Usage: The reversible nature of metamaterial deformations contributes to significant reductions in energy and material usage for repair and re-production with net-zero vision.

Lightweighting with Latticing: The inherent lattice structure of metamaterials provides an inherent advantage in lightweighting. Reduced material usage without compromising strength is a hallmark of these designs.

Vibration Control: The metamaterials with near-zero stiffness characteristics can be leveraged for band gap [4] and passive vibration isolation [5] applications. This feature enhances their utility in applications such as transportation, submarine technology, aviation, and space exploration, where vibration control/filtration is paramount for optimizing operational efficiency and ensuring safety.

Thermo-Mechanical Operation without Electronics: One distinctive feature of these metamaterials is their ability to function autonomously, relying purely on thermo-mechanical principles, without the need for complex electronic components. This opens doors to energy-efficient devices and systems that utilize energy scavenging processes as power sources.

Net-zero Manufacturing: 4D printing, when combined with these materials and designs, embodies a sustainable additive manufacturing paradigm. The use of bio-based materials minimizes their waste and ecological impact, promotes eco-friendly practices, and aligns with principles of sustainability. They offer the potential for biodegradation and recycling, achieving the goals of net-zero manufacturing and circular economy.

Lower Environmental Footprint: The advantages of reduced materials, shorter production time, and efficient energy usage contribute to lower environmental impact. These materials are aligned with sustainability goals, including the reduction of greenhouse gas emissions.

Diverse Applications: The transformative potential of 4D printing with mechanical metamaterials extends across an array of industries (see Figure 2), such as automotive/marine (e.g., energy absorbing bumper/fender/bonnet, suspension system/seat with vibration control), aviation (e.g., drone landing gear, wings, ani-vibration camera mount), protective gear (body armour, playground/care home flooring, safety helmet).

Figure 2. Potential applications for 4D printed metamaterials with shape memory and energy absorption/dissipation features for a resilient and sustainable future.

Current and Future Challenges

The field of 4D printing of shape-memory energy absorbing metamaterials presents a host of exciting opportunities, but it also faces significant technical challenges and limitations that warrant careful consideration. These challenges detailed below stem from the intricate nature of the technology and the ambitious goals it seeks to achieve.

Material Development: While progress has been made in developing bio-based materials for 4D printing, further research is required to enhance material properties. The challenge lies in striking a balance between sustainability and performance. Researchers must continue to explore new materials that are not only eco-friendly but also exhibit the required properties such as fast controllable activation, high shape-memory level, supreme strength, and excellent fatigue life under cyclic loading.

Reliability and Durability: Ensuring the reliability and long-term durability of 4D-printed products is crucial and remains a challenge. Variability in printed parts, defects, and material inconsistencies can affect thermo-mechanical and shape-memory performance. Controlling the degradation of materials and thermo-mechanical properties over time is critical. Developing robust quality control processes and standards is essential to build trust in the technology.

Scalability: Scaling up 4D printing processes for industrial applications remains a challenge. Achieving consistency, precision, and efficiency in large-scale production is essential for widespread adoption. Researchers and manufacturers need to overcome hurdles related to printing speed, size limitations, and material distribution on a larger scale.

Interdisciplinary Collaboration: Metamaterial 4D printing bridges multiple disciplines, including smart materials science, topological lattice design, advanced computational modelling, and additive manufacturing. Bridging the gaps between these disciplines and fostering effective interdisciplinary communication is a persistent challenge.

Accessibility and Expertise: The industrial adoption of 4D printing requires a skilled workforce capable of designing, programming, and operating the technology. Currently, expertise in 4D printing is concentrated in specialized research and development centres and universities. Expanding the accessibility of this technology and training a skilled workforce is crucial for its widespread adoption and harnessing its full potential.

Environmental Impact Assessment: As 4D printing gains traction, assessing its environmental impact becomes critical. Ensuring that the materials used in 4D-printed objects are truly recyclable and have minimal ecological impact throughout their lifecycle is an ongoing challenge. Evaluating the entire lifecycle of 4D-printed metamaterials, from material production to disposal, is essential to ensure that the technology aligns with sustainability goals.

Integration with Other Technologies: To fully unlock the potential of 4D printing, it must seamlessly integrate with other emerging technologies such as the Internet of Things (IoT) and data analytics. Developing compatible systems, artificial intelligence, and protocols poses a unique set of challenges.

Advances in Science and Technology to Meet Challenges

The challenges and opportunities presented by 4D printing of shape-memory energy absorbing metamaterials have spurred exciting developments in science and technology. Addressing these challenges requires a multifaceted approach, involving innovation across various domains detailed below:

Material Science Breakthroughs: A pivotal area of advancement lies in material science. Researchers are diligently working on developing novel bio-based materials [12, 13] that possess superior shape-memory characteristics while remaining environmentally sustainable. Expectations are high for materials with excellent printability, circularity, rapid activation, exceptional strength, and fatigue resistance.

Advanced Quality Control and Reliability Systems: Robust quality control mechanisms [14] are essential to ensure the reliability and durability of 4D-printed metamaterials. This involves the development of automated inspection systems, real-time monitoring of thermo-mechanical properties, and machine learning algorithms to detect defects and inconsistencies during printing.
Scaling Up Production: Researchers and manufacturers are exploring ways to overcome the challenges of scaling up 4D printing for industrial applications. Innovations in large-scale 3D printers, optimized printing processes, and precise material distribution methods are in progress to make mass production feasible [15].
Interdisciplinary Collaboration: To address the interdisciplinary nature of metamaterials 4D printing [16], institutions are fostering closer collaboration between smart materials scientists, metamaterials designers, computational modelling experts, artificial intelligence programmers, and additive manufacturing specialists. Cross-disciplinary research hubs are required to bridge these gaps effectively.
Empowering Access: Researchers are enhancing accessibility and expertise in 4D printing through user-friendly interfaces, simplified design tools, and comprehensive training programs [17]. Providing open-source platforms, and community-driven forums and promoting diversity and inclusivity within the 4D printing community could further enhance accessibility.
Environmental Impact Assessment Tools: Researchers are working on comprehensive tools to evaluate the environmental impact of 4D printing processes [18, 19]. These tools will analyse the entire lifecycle of 4D-printed products, from material production to disposal, to ensure alignment with sustainability goals.
Integration with Emerging Technologies: To fully harness the potential of 4D printing, efforts are being made to seamlessly integrate it with other emerging technologies like IoT [20] and data analytics [21]. This will create new opportunities for smart and adaptive systems.

Concluding Remarks

In conclusion, 4D printing of shape-memory energy-absorbing metamaterials embodies a journey towards innovation, sustainability, and transformative resilience. This cutting-edge technology, with its remarkable shape recovery, energy absorption, and adaptability, holds the potential to revolutionize multiple industries, including automotive/marine, aviation/aerospace, protective gear, and sports equipment. While facing technical challenges, ongoing research and interdisciplinary collaboration are driving advances in smart material science, quality control, scalability, accessibility, and integration with emerging technologies. As this field evolves, it brings us closer to a greener, more efficient, and eco-conscious future, harnessing the power of metamaterials and net-zero manufacturing for a resilient and sustainable world.

Acknowledgement

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3 – Shape Memory Polymer Roles in 4D Printing

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Status

Shape memory polymers (SMPs) are a type of intelligent material capable of recovering their initial shape from temporary deformation under external stimuli such as heat, magnetism, light, humidity, or pH changes. 4D printing can be defined as "3D printing + time", where complex shapes created through additive manufacturing techniques respond to external stimuli to exhibit changes in performance and functionality. The core of 4D printing technology is a programmable structure, and it cannot be realized without two important bases: mature additive manufacturing technologies and shape memory polymers that meet the functional requirements of 4D printing. The goal of 4D printing is to directly manufacture the structure with certain intelligent functions, which reduces the complexity and weight of the structures.

In recent years, 4D printing technology has made considerable progress in the field of medical devices. Wei et al. [1] developed a deformable and remotely actuated vascular stent using direct writing printing technology. The stent was implanted in the diseased part through minimally invasive surgery after it was given a temporary shape, and the magnetic field was applied to make the stent unfold to cure the vascular obstruction. Since then, 4D-printed medical stents have become research and development hotspots, such as cardiac stents, tracheal stents, urethral stents, sinus stents, orbital stents, and bone tissue [2-7]. Although the stents are not yet ready for clinical testing, researchers believe 4D-printed stents could be a steppingstone to more minimally invasive surgery in the future.

With the progress of the times and the development of science and technology, 4D printing technology is gradually entering our field of vision. Although 4D printing technology is still in its initial stages, its application prospect in biomedical, aerospace [8], clothing, furniture, and other industries cannot be ignored, as shown in Figure 1. 4D-printed deformable structures can make the aircraft better adapt to different flight environments reducing wind resistance and improving performance and efficiency. In addition, 4D-printed materials could help develop lightweight and flexible structures for satellites and spacecraft, reducing the cost and difficulty of space exploration. Using the deformation system, designers will be able to create clothing and accessories that can adapt to changing circumstances and meet the wearer's needs. 4D printing makes the self-assembly of furniture a reality. Consumers do not need to go through the tedious installation steps and tools but only observe the furniture self-formation process.

Figure 1. Applications of 4D printing of SMPs.

Current and Future Challenges

Although 4D printing is a manufacturing technology rising in recent years, it has faced a series of development bottlenecks, such as equipment, materials, and technology. How to overcome the constraints to achieve a breakthrough in 4D printing is the main task. 3D printers are the breakthrough point of 4D printing technologies. When we need to print large structures, we must use large machines to make large structures, so the spread and popularity of 3D printing technology are limited. To solve this problem, 4D printing has taken another path, relying on smart materials to give the printing structure self-deformation ability, which alleviates the limitations of the printer with the development of technology. However, intelligent printing brings higher technical requirements to the printing equipment, which will inevitably lead to the excessive cost of the equipment. In addition, printing accuracy is also a factor restricting the rapid development of 4D printing [9,10].

In addition to the constraints of the device itself, another problem with 4D printing is the special printing materials. 4D printing needs not ordinary materials but smart materials with memory function, which is a new functional material that can sense external stimuli and self-transform and assemble by judgment. At present, most 4D printing materials can only sense thermal stimulation. With the development of science and technology, the future is bound to realize the use of light, electricity, sound, magnetism, water, air, temperature, and other arbitrary trigger media, the trigger of self-deformation of material.

The development of new smart materials for different applications is the basis of the development of 4D printing technology. For example, materials for smart clothing should be soft and comfortable, and materials for deployable structures should be resistant to space environments. 4D printed implantable stents are used to treat luminal stenosis, whose slow recovery rate and low resilience are the main factors that restrict its development in biomedical field. In brief, the major challenges of 4D printing are developing new materials, controlling the recovery precision, speeding up the recovery speed and increasing the recovery power.

Advances in Science and Technology to Meet Challenges

Due to the influence of 3D printers, 3D printing materials, and technology maturity, 4D printing is still at the stage of research, development, and testing. It has no possibility of large-scale application, but the relevant research institutions worldwide actively layout, parallel follow-up. "While 4D printing is still in the experimental stage, it is a disruptive technology that will create a 'programmable world' that will revolutionize traditional manufacturing. The development of 4D printing is a dilemma that needs a breakthrough from many disciplines. First, high-resolution, high-speed, and multi-material 3D printing technology is required to rapidly produce multi-scale complex geometric shapes of materials and equipment. Secondly, the new multi-functional ink allows multi-functional materials to be used in 4D printing for further expansion. Finally, a theoretical model and design method are needed to predict and optimize the shape displacement accurately.

In the next few years, 4D printing is expected to make a breakthrough in the direction of new materials with high performance and an innovative technology of high-efficiency molding. 4D-printed shape memory polymers and intelligent structures have excellent prospects in the fields of aviation, aerospace, biomedicine, intelligent robots, intelligent homes, and intelligent clothing. The future will undoubtedly play an essential role in more industries and fields.

The development of 4D printing requires a collaborative effort among academia, industry, and government, each playing a positive role. The academic sector is responsible for knowledge transfer, talent development, and basic research; the industrial sector is responsible for transforming and applying research results to achieve productivity; and the government is responsible for direction guidance and policy support. No single university has all the answers, no single company sets all the standards, and no single country controls all the technology. The exchange and cooperation among academia, industry, and government accelerate the development and industrialization of 4D printing technology.

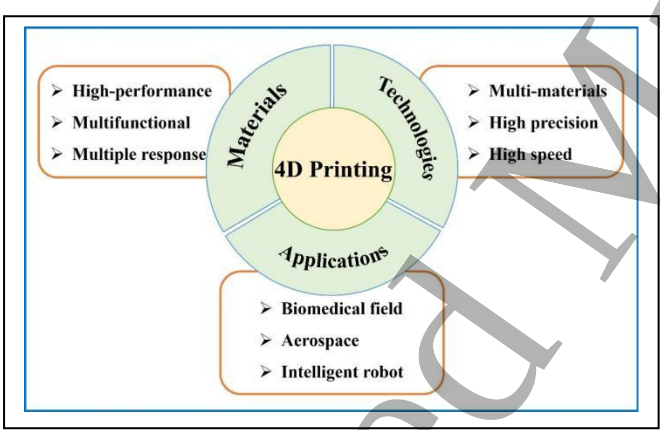

Figure 2. Future development direction of 4D printing.

Concluding Remarks

4D printing is an emerging manufacturing technology. Although there are many different printing methods, printable smart materials, and driving methods, 4D printing still faces many challenges in practical engineering applications. We need breakthroughs in new printing technologies, new smart materials, and new structural design and modeling software to promote 4D printing in soft robotics, biomedicine, aerospace, and intelligent electronic equipment, such as practical applications. Focusing on the transformation of manufacturing over the next decade, 20 years, and beyond, 4D printing is a forward-looking deployment of the country's major innovative technology. To encourage research-based universities and research institutes to actively follow up on the latest research progress in 4D printing technology and to conduct theoretical and basic research in materials science, biological sciences, industrial design, information coding, etc., the government encourages high-tech enterprises with research and development conditions to cooperate with research institutes to carry out experimental research and development of 4D printed products.

Acknowledgments

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4 - Liquid Metal 4D Printing

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Status

Liquid metals (LMs) refer to metallic elements or alloys that exist in a liquid state at or around room temperature. Gallium (Ga) is a popular LM because it has low toxicity and negligible vapor pressure. The melting point (~30 °C) of Ga can be depressed below room temperature by alloying it with other metals such as indium, tin, or zinc. Ga and its alloys find use as soft metallic conductors in electronics, soft robotics, and thermal management systems.

One of the most important features of Ga-based LMs is the formation of a thin (1-5 nm) metal oxide layer that occurs spontaneously on the surface when exposed to air. The oxide is a solid shell that stabilizes non-spherical liquid structures, such as cylinders or cones, that would otherwise be unfavorable due to the extremely high surface tension (640 mN/m) of bare LMs [1]. The oxide allows LMs to be printed [2]. In essence, the liquid-like nature allows the LM to be extruded from a nozzle, while the unique rheological properties provided by surface oxides allow it to maintain its shape once printed [3]. In addition, LM can be mixed, integrated, or blended with polymers to create other types of printable inks [4].

4D printing refers to changes (mostly shape morphing) of 3D-printed objects in response to external stimuli. Currently, materials used for 4D printing are generally specialized polymers that do not conduct electricity. Metallic materials are interesting for 4D printing because they allow for the fabrication of dynamic conductive parts and circuits.

LMs are appealing for 4D printing because they can be dispensed at ambient temperature. Yet, there are only a few examples that use LM for 4D printing. Figure 1 shows a recent example of a printable 'metallic gel' composed of Cu particles connected by LM particles in water. After printing, the part changes shape as it dries due to stress arising from a rheological modifier (polymer) added to the ink [5].

To date, most efforts to 4D print LMs are achieved by combining LM with other materials, such as polymeric materials, that can generate stress post-printing. The printing has mostly been done via direct-ink-writing (DIW) technique. This field is in its infancy, yet the prospect is fascinating. 4D printing of such LM-containing materials can create parts with metallic electrical and thermal conductivity in addition to other unique properties, such as stretchability and self-healing properties – features that are difficult to achieve with conventional solid metallic materials.

Current and Future Challenges

LM 4D printing can be broadly defined as all 4D printing materials that incorporate LMs. Such materials should be printable and have a dynamic response (e.g., shape change) to a stimulus applied to the part post-printing. 4D printing often utilizes stored stress in the initial 3D printed part. When stimulated using heat or light, the stress releases to cause a shape change. Yet, bulk LM is a low viscosity fluid and therefore cannot store bulk stress, which is a challenge for directly 4D printing. However, LMs do have some unique features that can be harnessed for 4D printing. For example, bare LMs have large surface tension that can be electrochemically modulated [6], which could be used to drive shape change. In addition, the oxide layer allows LM particles in inks to be sheared into aligned, elongated shapes during printing. Such shapes can cause anisotropic shrinkage that can be harnessed for 4D printing [5].

In addition, LMs can be rendered into responsive composites by either distributing LMs into a responsive matrix (e.g., polymer) or by adding fillers to the LM to make it responsive (e.g., magnetic particles) [7].

Figure 2 shows that LMs participate in 4D printing primarily in three roles: 1) As non-active fillers. In such cases, the response of the printed parts is governed by the matrix material, such as polymer. The role of LM is to impart specific functionalities to the printed components, like conductivity, without adding stiffness; 2) As indirectly active fillers (cf. Figure 1). In such cases, the presence of LM can either alter the responsive traits of the matrix or confer responsiveness upon otherwise non-responsive materials; 3) As active components. In such cases, 4D printing is realized in the printed material through the intrinsic response of LMs to stimuli.

The primary challenge in LM 4D printing of composites is to find the right trade-off between printability and functionality. Poorly designed processes could result not only in the forfeiture of specific properties (e.g., conductivity) but also in the deterioration of the shape morphing properties of printed parts.

The other challenge is determining the right source of stress to cause a shape change. Current LM 4D printing methods primarily rely on responsive polymers to trigger shape change, yet LMs have a variety of routes to become electro-responsive actuators, which suggests new routes for future 4D printing based on LM [8].

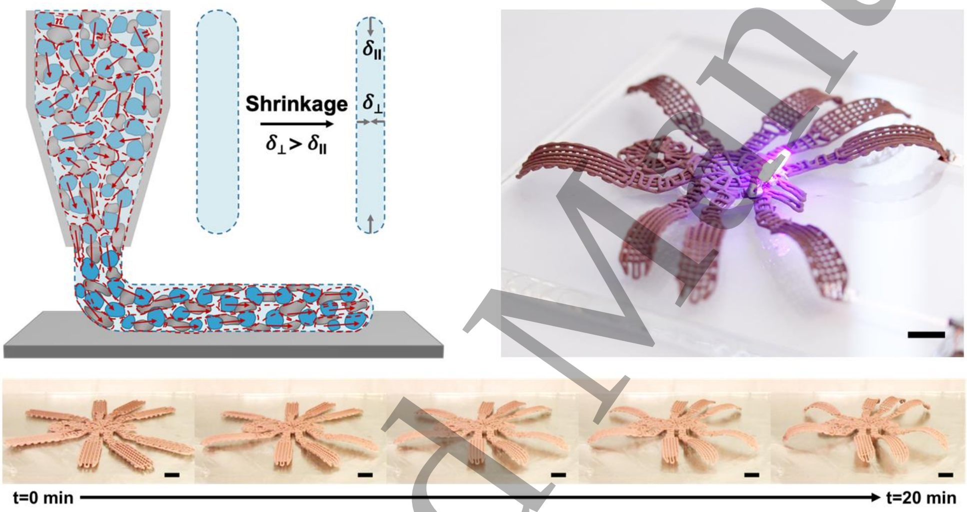

Figure 1. Example of a 4D printed liquid metal/copper composite with shape morphing characteristic. Reprint from [5], Copyright (2023), with permission from Elsevier.

Advances in Science and Technology to Meet Challenges

There are several ways to print LMs, as reviewed recently [1]. One challenge is that LMs naturally form droplets if extruded from a nozzle. This issue can be overcome by 'shearing' the LM from a nozzle by moving a meniscus of the LM across a substrate [9]. Another approach is to modify the rheology of the LM by adding particle fillers [10]. A third approach is to encase the LM as it exits the nozzle in a core-sheath filament [11]. A fourth approach is to distribute LM in a composite or polymer matrix. [4] These LM printing techniques can inspire LM 4D printing strategies.

Since bulk LM is a low viscosity liquid, one of the challenges is finding ways to generate stress in parts to promote shape change for 4D printing. One approach is to simply incorporate LM with active dopants that respond to external stimuli. For example, coaxially printing LM with a silicone shell containing magnetic particles (Figure 2A) produces magnetic responsive parts while the high conductivity of the LM confers sensor functionality [12]. Another approach is to indirectly use LM to aid 4D printing. In one example, a metallic 'gel' consisting of Cu particles connected by LM particles contains up to 97.5% metal, making it highly electrically conductive (Figure 2B) [5]. The wetting of LM to the Cu leads to the formation of an elastic network of particles. Extruding the ink through a nozzle shear-aligns the LM into elongated particles that impart anisotropic shrinkage during drying of the printed part to enable 4D printing. In this example, the stress arises from a small amount of polymer added to the ink. Another approach is to harness the conductive of LM to create electromagnetic forces controlled by electrical current (Figure 2C) [13]. Phase transitions of LM can also affect the mechanical properties of LM/polymer composite, thus providing a way to stiffen or soften 4D printed polymers.

There are promising routes by which LM might generate stress. For example, electrochemical reactions (Figure 2D) or electrocapillary effects (Figure 2E) can modulate surface tension and thus drive actuation [14-16]. LM can also be used as a component in an electromagnet to generate force [17]. Furthermore, LM expands when it freezes, which can generate stress due to volume changes. By coupling such mechanisms with suitable printing designs, there should be multiple routes to 4D print LM.

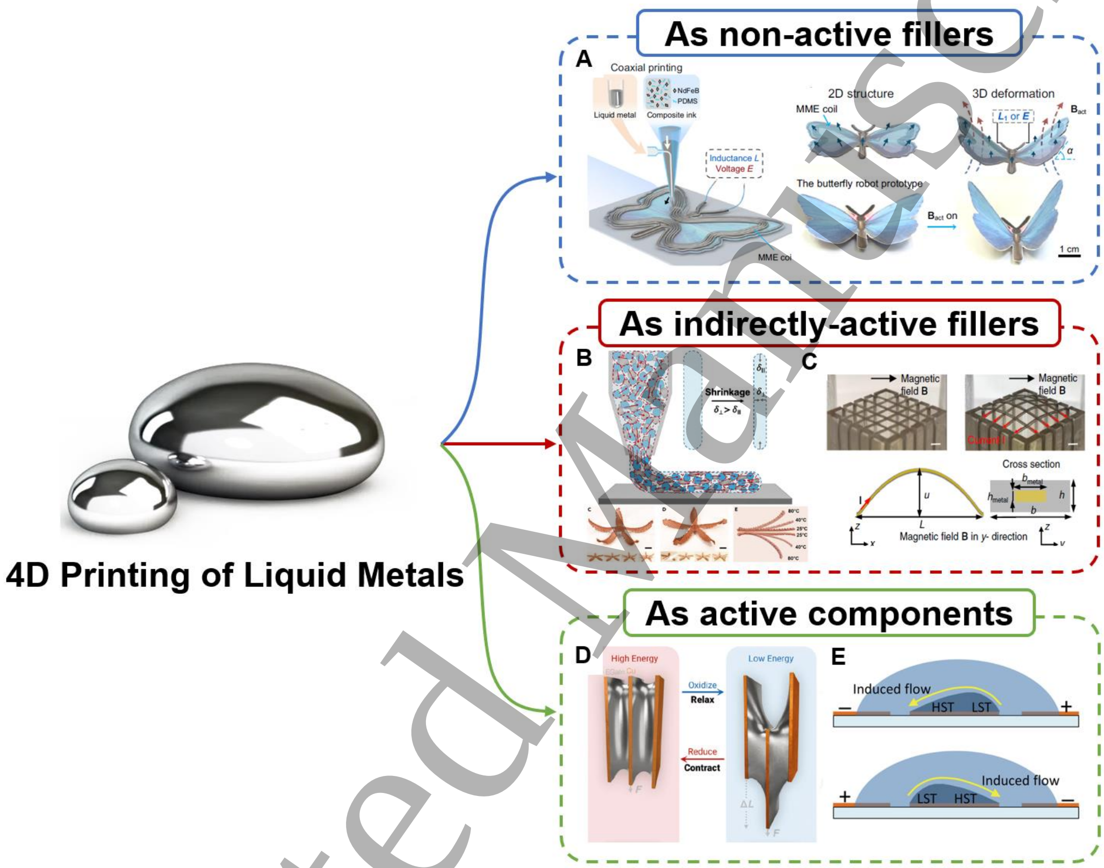

Figure 2. The role of LMs in the 4D printing process. A) LMs are printed along with 4D printing materials as nonactive fillers. Reprinted from [12], Copyright (2023), with permission from Springer Nature. B-C) LMs are affecting the 4D printing process together with the printing materials. Reprinted from [5], Copyright (2023), with permission from Elsevier; [13], Copyright (2023), with permission from Springer Nature. D-E) LMs act as the stimulus responsive components in the 4D printed parts. Reprinted from [14], Copyright (2022), with permission from Wiley-VCH; [15], Copyright (2014), with permission from Wiley-VCH. Copyright (2014), with permission from Wiley-VCH.

Concluding Remarks

Despite being in an early stage of research, the 4D printing of LMs is poised to attract substantial interest because of the ability to (1) print such materials at low temperatures, (2) create parts with metallic properties, and (3) enable unique routes for generating the stress necessary for 4D printing. 4D printing of LMs may enable applications ranging from dynamic electronics to thermal interface materials and soft robotics. To reach these goals, it is necessary to (1) dedicate further efforts to exploring more efficient LM patterning technologies; (2) explore ways to harness the attributes of LM for 4D printing;

(3) apply the actuation principles of LMs, such as surface tension modulation - to fit the current LM 3D printing technology.

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5 - 4D Printing of NiTi-based Shape Memory Alloys

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Status

Shape memory alloys (SMAs) are a group of smart materials distinguished by their unique properties, namely the shape memory effect and pseudoelasticity These properties stem from the reversible transformation between two main phases, austenite and martensite, with different structures. Phase transformation is induced by either heating or loading the material, and it can be reversed upon cooling or unloading, respectively [1, 2]. These characteristics bring about the application of SMAs in various fields, such as biomedical devices, aerospace, and actuators. NiTi is the most studied and commercially available shape memory alloy and its fabrication through additive manufacturing has been extensively investigated over the past decades (2). 4D printing of NiTi-based SMAs is gaining interest in further developing the material for automotive, sensors, and aerospace applications [3, 4]. A 4D-printed bending composite with NiTI wires as filament is shown in Figure 1. 4D printing involves the behavior change of the material in a defined way over time, which is stimulated by an external source such as heating. The self-repairing benefit of 4D printing is a revolutionary feature, especially for aerospace applications. Nevertheless, further investigation, including development in 3D printing techniques, is required to improve 4D printing to produce functional parts [5, 6].

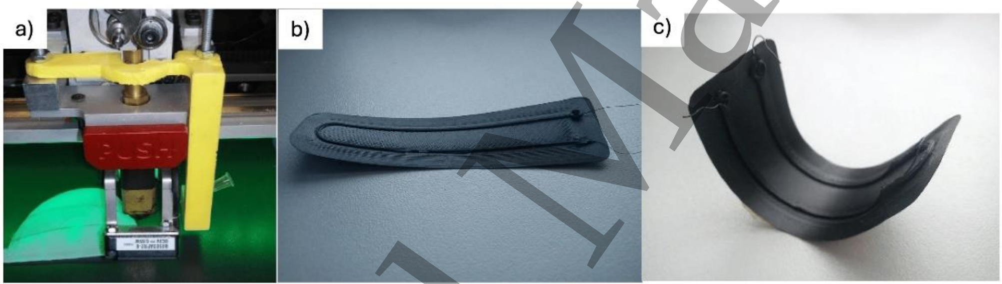

Figure 1: a) FDM additive manufacturing process used to produce actuators with NiTi filaments, the printed actuator b) before and c) after activation, obtained from [7].

Among various methods, 4D printing of metallic components has been mostly investigated through the powder bed fusion (PBF) and direct energy deposition (DED) processes.In a study by Hassanin et al. [8], they were able to design and fabricate re-entrant structures using NiTi with improved ballistic performance. In the field of actuators, 4D printing benefits of NiTi-based SMAs were investigated by Sreesha et al. [9] on NiTi, NiTiCu and NiTiFe wires , focusing on the debonding strength of the matrix-alloy interface, mechanical and functional properties. Among the investigated alloys in this study, NiTiFe exhibited the highest debonding strength and lower strain. NiTiCu actuators had a larger number of actuation cycles and NiTi actuators obtained higher strain recovery. Zhan et al. [10] developed 4D printed NiTi parts with gradually graded transformation behavior with 7.4% elongation. Chen et al. investigated 4D printing of equiatomic NiTi thin walls on TC4 substrate by electron beam DED method [11]. They obtained graded microstructure with different Ni-rich or Ti-rich precipitates at regions of the part along the height, which resulted in different mechanical behavior while the recoverable strain was relatively constant. It is important to consider that 4D printing of NiTi-based SMAs is at an early stage and more studies will be performed to benefit from its characteristics in the future. The development of the mentioned methods, and other metal additive manufacturing techniques, can revolutionize the industry and market for 4D-printed NiTi-based SMAs. These advancements can result in fabricated parts holding the advantages of self-assembly, self-adaptability, and self-repair associated with 4D printed materials [6].

Current and Future Challenges

4D printing has the advantage of producing more flexible and efficient parts, which leads to broader applications compared to 3D printing. Adaptability to the environmental conditions and self-improving of the material's properties and behavior are the unique features of 4D-printed structures compared to conventional fabrication methods [6]. However, there are challenges in this field regarding the material and the implemented technique. In terms of technology, the limited availability of metal 3D printers with the capability of 4D printing is a drawback to further investigating and developing NiTi-based SMAs processing. Currently, the powder bed fusion technique is the most studied process by researchers since this method is well investigated and developed for 3D printing compared to the other techniques; direct energy deposition has also been investigated in a few studies. Implementing other methods and techniques, such as binder jetting, can help researchers benefit from their advantages. Besides, the mentioned processes (PBF and DED) are at the primitive stage of investigation and needs further development. Developingmore technologies also helps to use other forms of feedstock, such as wire. The use of other techniques also enables the potential of *in situ* alloy design to significantly improve the properties. This has been proven to improve the mechanical properties of NiTi with the laser DED process by implementing Ni-rich nanocomposites in the microstructure [12]. This can be studied in 4D printing and investigate its functionality. The very properties of NiTi responsible for the advantages of 4D printing, i.e., the shape memory effect and pseudoelasticity , bring about challenges in their manufacturing. The composition sensitivity of the alloy that directly affects its properties needs to be carefully monitored in 3D printing due to the rapid solidification nature of the powder bed fusion method. The presence of impurities introduced during fabrication can alter properties such as transformation temperature, and bring about precipitates that are detrimental to the mechanical performance and functionality of the part [6]. These are also relevant challenges in 4D printing and need to be studied.

Advances in Science and Technology to Meet Challenges

4D printing promises unique and revolutionary properties; however, it still needs to be studied further to implement more technologies. 4D printing of NiTi-based SMAs can advance in terms of available methods of fabrication, alloy design of the material, and developing NiTi-based alloys to benefit from other elements properties to enhance performance, leading to an increase in the potential applications of NiTi 4D-printed structures [6]. To date, 3D printing of NiTi-based shape memory alloys has been extensively studied. Elahinia group at the University of Toledo [13-15] have performed a comprehensive study on the effect of process parameters of laser powder bed fusion (LPBF), namely laser power, scanning speed, and hatch spacing, on the microstructure and mechanical properties of NiTi SMAs on the resulting microstructure and texture is also investigated to optimize process parameters to be able to print dense parts with improved mechanical properties. In these studies, the relation between processing and properties is also coupled with post processing, such as hot isostatic pressing and heat treatment, to investigate the benefits of post processing to further improve the properties and performance by removing the cracks or by the formation of the precipitates that are favorable for mechanical performance [16, 17]. They were also able to design and print functionally graded NiTi that can further improve the technology by eliminating the need to joint or weld [18]. Numerical studies and modeling are effective ways to reduce the associated costs with experimental studies. Predicting properties such as melt pool dimension and morphology [19] and the effect of hatch spacing on superelasticity [20] in laser powder bed fusion has been studied.

Current challenges in 3D printing of NiTi-based SMAs, such as controlling composition, transformation temperatures, and residual stress, are applied to 4D printing as well. Therefore. further studies in terms of technology and alloy design are essential [2]. The self-repairing and self-enhancing properties of 4D-printed parts are cost-effective due to less material consumption and the elimination of repair costs [6]. This is especially beneficial for aerospace structures and, overall, can significantly affect the industry. Overcoming these challenges can help further benefit from the potential of NiTi-based SMAs and improve its application in aerostructures, actuators andbiomedical devices. The flowchart in Figure 2 summarizes the challenges and the potential improvement of NiTi 4D printing in terms of material and technology. To advance this technique, collaboration between academia and industry is essential. The collaborative studies between different groups of material scientists, engineers and manufacturers can accelerate the pace of this technique to the next stages of development and real-time applications. Another important field in 4D printing is modeling to stimulate the performance of the designed part. Another area is testing prototypes to observe if the part shows the expected functionality and performance. Also, seminars and workshops on the benefits of this method can attract researchers and industry to investigate this field.

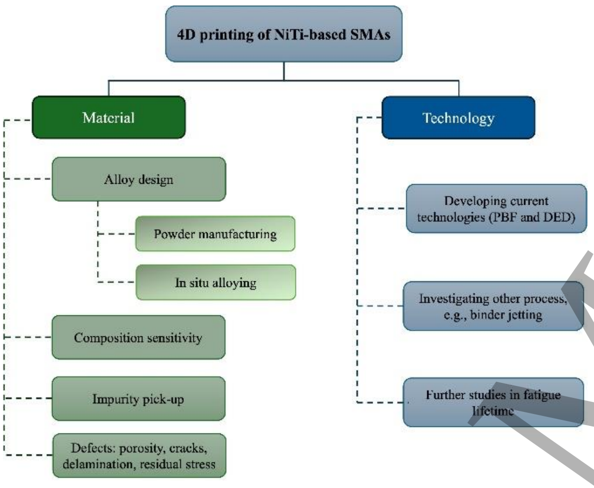

Figure 2: A flowchart illustrating the challenges and prospects to improve 4D printing of NiTi-based shape memory alloys within the realm of material processing and technologies.

Concluding Remarks

4D printing of NiTi-based SMAs is at its initial stage of study and development. It can be advanced by combining complementary studies on the current methods and introducing new techniques to benefit from their advantages in terms of feedstock form, *in situ* material design, and resulting properties. Development in this area also helps to study other NiTi based shape memory alloys and benefits from coupling NiTi with other elements such as Hf, Cu, Fe, and Mn. An important field in 4D printing is modelling, which can provide a prediction of the performance of the designed part over time. Incorporating more studies in this area can also help to purposefully alter material composition and processing optimization. Overcoming these challenges and broadening the study of 4D printing of NiTi-based SMAs can revolutionize the application of these materials, especially in aerospace structures where they can benefit from the self-improving and self-repairing features of the manufactured parts.

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6– 4D Printing of Composites

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Status

4D printing of composites (4DPC) was first conceived by Hoa in 2017 [1]. This technique provides a method to manufacture curved, structural, components made of composite materials having long continuous fibers embedded in a polymer matrix, without the need for a curved mold. As such, the term of “Moldless composite manufacturing” is also used to refer to this technology. These are the same materials that have been used to make structural components of aircrafts, automobiles, wind turbine blades, and high-end sports equipment. These materials have high strength (about 1000 MPa) and high modulus (about 180 GPa). The principle of operation of the technique depends on anisotropy. Figure 1 (left) shows a schematic of a sheet of prepregs. Within each sheet, the fibers are aligned along a certain direction. The material properties along the fiber direction are different from those transverse to the fiber direction. The prepregs consist of fibers embedded in uncured thermoset resin (such as epoxy). The uncured resin is a flexible, and sticky, solid. To manufacture a component with curved shape, thin sheets (or strips) of the prepregs of the composite are stacked one on top of the other, on a flat mold. Figure 1 (right) shows a stack of two sheets, one with fiber along the 0^o direction, and the other along the 90^o direction. The thickness of each sheet is about 0.125 mm. Due to the softness, and stickiness, of the uncured resin, the two sheets do not interact with each other (except sticking to each other) when the stack is first deposited. The stack of prepregs is then placed in either an oven or an autoclave to be cured at an elevated temperature. The curing hardens the resin. When the cured laminate is cooled to room temperature, mechanical interaction between the sheets, due to the differences in the coefficients of thermal expansion of the sheets in different directions, causes the flat stack to become curved. By proper control of the fiber orientation in each layer in the stack of prepregs, one can obtain structural components of different shapes. The mechanism responsible for the shape change is the anisotropy of the laminate. This anisotropy comes from the differences in the moduli and coefficients of thermal expansion (or contraction) along the fiber direction, and transverse to the fiber driecton. This technique has been used to successfully manufacture composite pieces of S shapes [1], composite leaf springs [2], letters of the alphabet [3], twisted pieces [4], composite flower, Omega stiffener [5], corrugated core for flexible aircraft wings [6], and conical shells [7]. One particular example is the case of composite leaf springs [2] (figure 2). A 60-layer laminate with lay-up sequence of [$90_{36}/0_{24}$] was used to make curved springs having a stiffness constant of 486 N/cm. The springs were subjected to fatigue loading in which the spring was alternately loaded to be flattened, and unloaded to become curved, more than 1 million times without any sign of degradation.

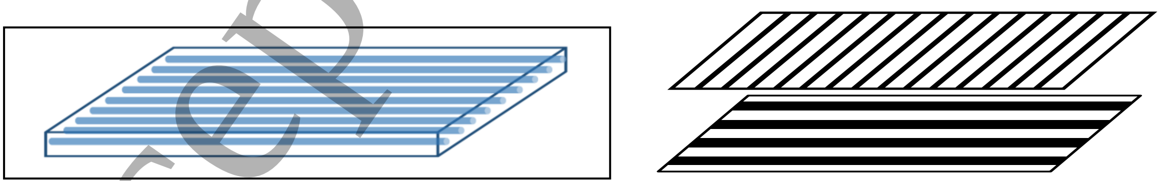

Figure 1: (Left) Unidirectional fibers in a single sheet of prepreg. (Right) A stack consisting of one layer with fibers along the 0^o direction and the other layer with fibers along the 90^o direction.

Figure 2: Composite leaf spring.

Current and Future Challenges

The technical challenges and limitations of 4DPC can be as follows:

1. 4DPC structures have been made using thermoset composites such as carbon/epoxy. For outer space applications, thermosets such as epoxy have outgassed problem. Thermoplastic composites would be preferred. However, processing thermoplastic composites using an automated fiber placement machine to make unsymmetric laminates would be a challenge. The reason is due to the instantaneous solidification and softening of the thermoplastic composites when the temperature changes from close-to-melting down to close-to-room temperature many times during the process. Significant temperature gradients exist in three dimensions during the process, and this significantly affect the shape transformation of the structure.
2. ***The effect of temperature and environmental moisture on the stability of the dimension of structures made using 4DPC***. Since the curved shape of composite structures made using 4DPC depends on temperature to attain it shape, the variation of temperature in turn can have effect on the shape during the operation of the structure. This issue needs to be addressed. There is a range of temperature and moisture where the effect is small and reasonable operation can be possible.
3. **It is a niche application**. Not all curved structures can be made using DPC, particularly thicker structures. Thinner structures can react more to anisotropy while in thicker structures the effect of anisotropy dampens out. However, the demarcation line is not clear. There have been new composite structures made by 4DPC that were thought to be impossible previously. There is a need to develop more composite structures using 4DPC.
4. ***4DPC uses an automated fibre placement machine to deposit the prepreg strips to make layers.*** Not all laboratories have an automated fibre placement machine. The reason is because the cost of the machine is high. The operation and maintenance costs are also high. With increasing use of automation in the manufacturing of composites, hopefully the cost of the machine can be decreasing. Also there have been development of smaller automated fibre placement machine with affordable costs.
5. ***The behavior of the unsymmetric laminate in operating conditions.*** Most work to study the behavior of composite laminates in operating conditions (mechanical loading, vibration etc.) have been done on symmetric laminates. There is a need to study the behavior of un-symmetric laminates in operating conditions.
6. There is a need to find ***industrial applications***.

Advances in Science and Technology to Meet Challenges

There is more and more interest from the younger researchers on the 4DPC technology. Many potential projects can be carried out:

1. ***Using 4DPC to make structures for outer space applications***. These structures can be used as housing for equipment that needs to operate during the day but needs to be insulated from the

cold during the evening. Structures made using 4DPC can open and close by itself, depending on whether it is day or night. During the day, the structure is heated by the sun. At higher temperature, the structure can open to allow the equipment to function. In the evening, the temperature decreases, the composite structure then closes itself, and insulates the equipment from the cold.

2. ***Composite structures used for efficient packaging***. In situation where it is necessary to send a package to a remote area (outer space, or to the territories close to the North Pole), it is more difficult and more costly to send the structure in 3D shape. 2D shape of the same volume of materials may be more efficient for packaging, and shipping. In this case, flat layers of prepregs may be laid down to make a flat stack. This is shipped to the remote location. On arrival, this stack of prepregs can be heated up to cure. It is then cooled down to ambient condition, to take up the 3D shape.
3. **There is more and more interest on the sustainability of materials and structures.** The use of 4DPC provides a means to make composite structures without a complex mold. This provides savings in cost and time. This can represent opportunities for better sustainability.
4. The applications of 4DPC to make components in the **automotive industry** where the requirements for perfect shape may not be very stringent.

Concluding Remarks

4DPC is a brand new technology. It provides the ability to make composite structures of complex shapes, containing long, continuous fibres, using only a flat mould. This gives significant savings in time and cost. These structures not only have the complex shape, they also exhibit high strength and stiffness, which make them suitable for load bearing applications. Structures that have been made using this technology include pieces with S shape, composite leaf springs, twisted laminates, omega stiffeners, corrugated core for flexible aircraft wings, and conical shells. There are also unique structures that only 4DPC can manufacture, such as composite flower, while conventional composite manufacturing using a mold of complex shape can not. This opens up many opportunities for new designs and new applications. The advances of this technology can have major impact on the effective utilization of resources, which has impact of the sustainability of the technology. While the technology opens up many exciting opportunities, there are challenges to overcome. These include the need to understand the behavior of unsymmetric laminates in operating condition, and the range of temperature for efficient operation.

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7 - Functionally Graded AM for Shape-changing 4D-textiles

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Status

The integration of additive manufacturing and textiles results in the hybrid material "4D textile," where the interaction between the flexible textile and the printed structure leads to a transformation from a 2D configuration to a 3D morphology. [1, 2] The textile stores potential energy by pre-stressing before printing. Upon releasing this pre-established tension after printing, it generates a restitutional force in the textile, countering the inherent elastic rigidity within the printed zones. Returning to an energy-favorable state of balance causes the change in shape from a two-dimensional printed structure to a spatial design. [1] The choice of materials for 4D textiles depends on various factors, such as the selected additive manufacturing process or the desired properties of the end product. Knitted and warp-knitted fabrics containing elastic yarns are used as substrates in 4D textiles because they have high elastic elongation and sufficient restoring force. Material properties of the textile must be considered when selecting the polymer and printing parameters to prevent damage to the textile. [1]

Nowadays, multi-material additive manufacturing (MMAF) using the fused filament fabrication (FFF) process is becoming increasingly significant. This is due to the continuously expanding selection of filaments with different physical, mechanical, and electronic properties. Additionally, the process parameters in the FFF process can be tailored more individually. [3] With multiple extruders, FFF allows for the extrusion of various materials. The number of nozzles is determined based on the process's requirements. Materials can be mixed within a single nozzle using a static or dynamic mixer to enhance the interface strength of the printed part [4]. Alternatively, materials can be extruded separately through multiple nozzles [5]. These advanced capabilities offer new perspectives and enable the production of intricate products with customized attributes. In conventional MMAF, the combination of two materials is limited to discrete interface areas, often leading to high stress concentrations and potential failure zones. Thus, the concept of functionally graded materials (FGMs) emerges [6].

While MMAF aims to blend different materials within an object, functional graded additive fabrication (FGAF) fine-tunes the properties of the material itself. FGMs exhibit a gradual change in volume ratio or physical properties within the material [7]. Their notable resistance to contact deformation, damage, and crack propagation is achieved through this gradual transition [8, 9]. This transition facilitates a skilled redistribution of stresses and reduces stress concentration at interfaces.

Current and Future Challenges

The synergy between FGAF and 4D textiles aims to enhance the mechanical properties of 4D textiles. PLA is often employed as a support material in 4D textiles. However, the resulting structures can be susceptible to breakage due to PLA's stiffness. To address this issue, soft TPU is incorporated using FGAF to impart flexibility and enhance structural durability.

The combination of soft and rigid materials through FGAF introduces multifunctionality to 4D textiles. This innovation allows for the creation of adaptive textiles capable of selectively altering their shape and properties without compromising structural integrity. These advancements hold promise for a range of applications, including wearables, medical technology, and robotics.

Nevertheless, several challenges must be addressed in the integration of FGAF and 4D textiles. Factors influencing adhesion between the textile and the structure, as well as the structural changes themselves, are numerous. Challenges also arise from the FGAF manufacturing process and the selection of suitable material combinations.

Optimizing adhesion between the printed structure and textile is paramount. The adhesion mechanism relies on the molten polymer infiltrating the pores of the pre-stressed textile during printing and solidifying during cooling. This forms a mechanical bond between the 3D-printed structure and the textile. However, the retraction of the pre-stressed structure can lead to detachment, primarily during bi-stable state transitions.

The shape change of 4D textiles is influenced by various factors, including the initial textile pre-stress, textile properties, and the geometry of the printed structure. The Institute of Textile Technology (ITA) has already conducted extensive research on the factors affecting shape changes and adhesion, with further analyses planned.

In the FFF process, precise control of the material ratio in each layer poses a challenge. In this study, a printer equipped with two extruders and one nozzle was used. The extruders can be individually controlled to tailor the material composition. However, variations in material distribution in the xy-plane can occur due to dead volumes in the nozzle and long Bowden tubes, resulting in printing delays. Enhancing printer stability and improving manufacturing process traceability are necessary to address these issues.

Additionally, not all materials can be easily combined in a single printing process, especially with a single-nozzle printer. Careful selection of nozzle temperature is required to ensure uniform extrusion of both materials and avoid potential problems such as under-extrusion or structural defects.

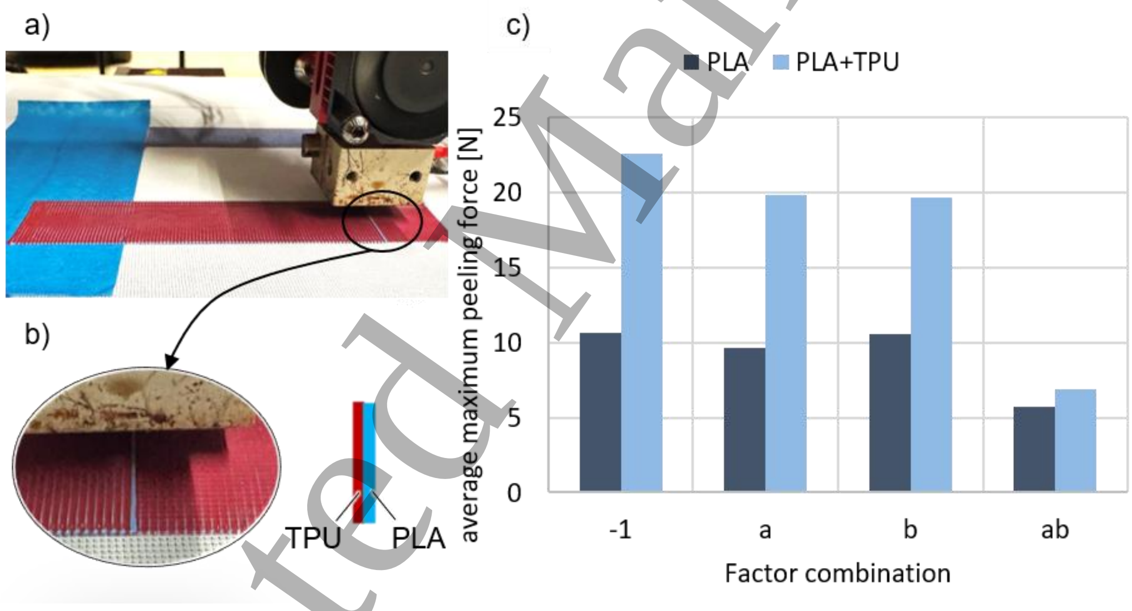

Figure 1. (a) Preparation of specimens for adhesion testing (b) an enlarged detail of the sample with a schematic representation showing the positions of TPU and PLA (c) Comparison of the average maximum peeling force of the samples from PLA and PLA/TPU mixture.

Advances in Science and Technology to Meet Challenges

To achieve success, 4D textiles must seamlessly integrate with existing materials and provide a deeper understanding of the process. Key advancements include improving the adhesion between printed components and textiles, analyzing shape changes, developing a predictive and reliable printing process, and exploring diverse material combinations and various structures.

This represents the inaugural application of Functionally Graded Materials (FGMs) in 4D textiles. To achieve this, apart from the manufacturing process, the development of interface adhesion and the analysis of shape changes in 4D textiles were crucial. This comprehensive approach effectively tackles the challenges and ensures the successful implementation of 4D textiles with FGMs. Adhesion serves as a critical criterion for the quality of 4D textiles and is influenced by factors like material texture, nozzle-textile distance, printing speed, and nozzle/print bed temperatures. In this context, a blend of PLA and TPU is printed on the textile to examine multi-material adhesion. In the blend, the volume ratio of PLA to TPU is 1:1, as depicted in Figure 1. These two materials are not homogeneously mixed but placed side by side. In this case, a Design of Experiments (DoE) was conducted. The diagram clearly illustrates that the adhesion of PLA and TPU is significantly higher than that of pure PLA samples, highlighting TPU's enhancing effect due to its lower viscosity, which aids penetration into textile pores and fosters multiple mechanical bonds.

The printed FGM sample exhibits pure PLA content at the left edge (normalized length 0) and pure TPU content at the right edge (normalized length 1), with the PLA content decreasing along the length while the TPU content increases, as seen in Figure 2. The gradient length is set at 70% of the total length, considering the printer's current maximum capability. Longer gradients could risk variations in material composition between layers, altering the visual transition.

Distinct mechanical properties of PLA and TPU result in different curvatures, as shown in Figure 2. Due to the edge effect, the corresponding curvature is considered between 0.1 and 0.9 of the normalized length. The TPU sample's lower strength leads to higher curvature compared to PLA. In FGM samples, curvature increases with the rising TPU volume fraction along the length, intersecting the TPU specimen curve at approximately 90% normalized length. This indicates near-complete TPU dominance and identical curvatures. Such insights could be pivotal in applications like soft robotics employing 4D textiles.

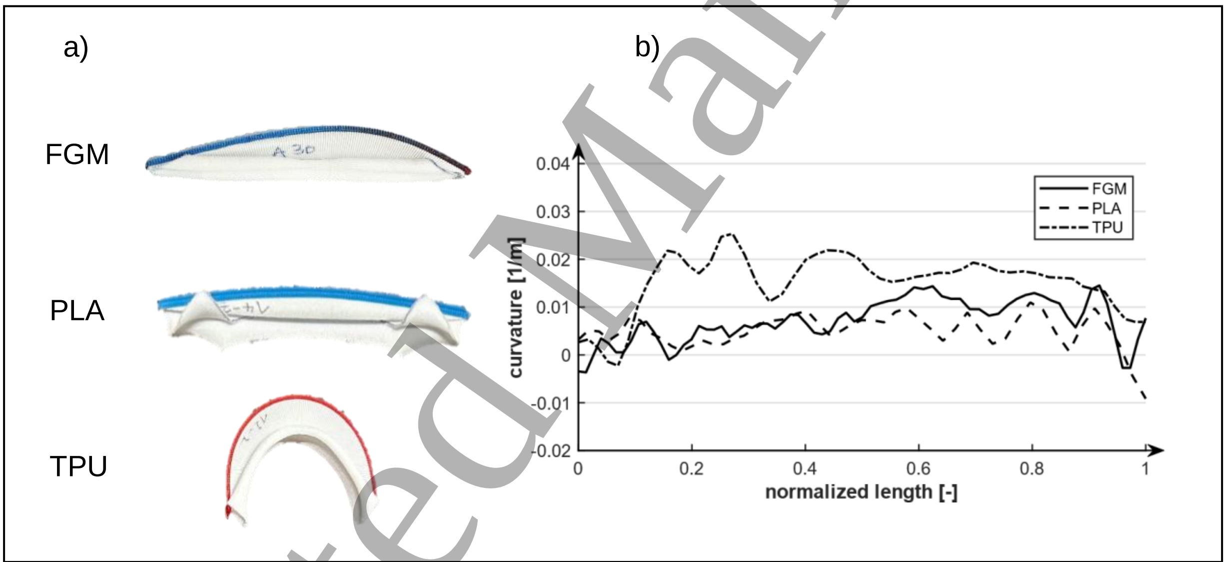

Figure 2. (a) Specimens made of different materials (b) Comparison of the curvature of the specimens along the length.

Concluding Remarks

The evolution of 3D printing for crafting 4D textiles is on the verge of significant advancements in the near future. Current efforts are focused on material and structural analysis, innovative manufacturing techniques, and novel material combinations. By surmounting technical challenges and enhancing material composition, these materials hold tremendous potential for applications like soft robotics. However, ongoing refinement of the printing process and the exploration of adaptable material combinations are essential to unlock the full capabilities of this technology.

The integration of Functionally Graded Materials (FGMs) into 4D textiles represents a substantial leap forward. By gradually altering the material composition within a printed part, it becomes possible to achieve specific mechanical properties in a targeted manner. This opens the door to the creation of custom structures that are both flexible and durable. Such materials could usher in revolutionary changes in fields like soft robotics, architecture, medicine, and wearables.

The development of precise printing processes and the continuous adaptation of material combinations are pivotal for harnessing this potential to its fullest extent. If these challenges can be successfully addressed, 4D textiles enhanced with FGMs have the potential to make a profound impact on various industries by offering innovative solutions to complex problems and unveiling new design possibilities.

Acknowledgements

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8- 4D Printing of Piezoelectrics

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Status

One of the most crucial materials for 4D printing is Piezoelectric materials due to their ability to convert electrical energy into mechanical deformation or vice versa. This property enables precise and programmable shape changes in printed objects over time, enhancing the dynamic capabilities and functionality of the printed structures in response to external stimuli on demand. Moreover, the precision of 4D printing has enabled intricate piezoelectric architectures, promising advancements in soft robotics and adaptive aerospace structures. Collectively, these breakthroughs highlight a paradigm shift towards intricate, responsive materials that merge the potential of 4D printing with the unique attributes of piezoelectricity.

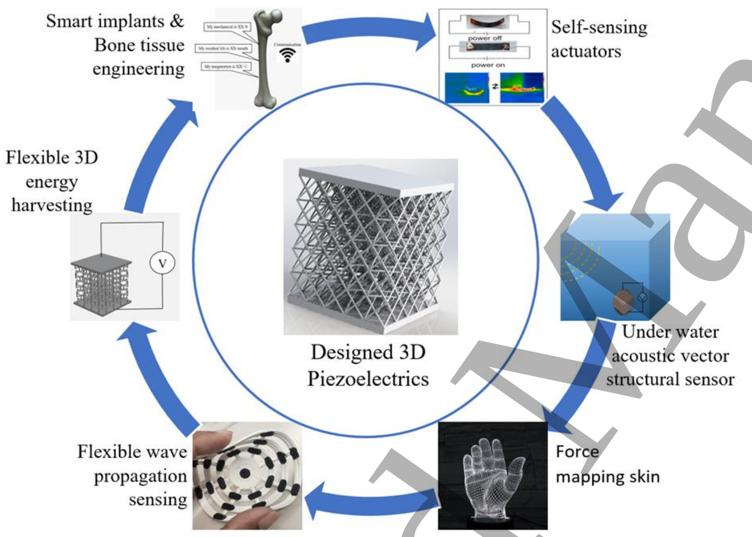

Figure 1: A visual representation outlining the transformative potential of intricate piezoelectric design and 3D printing across diverse domains [1]-[3]. Reproduced with permission.

From creating self-sensing actuators [1], self-transforming devices [2], and responsive sensors [3] to smart implants [4] and bone tissue engineering [5] in the medical sector, the applications of the 4D printing of piezoelectric materials are enormous. **Figure 1** visually depicts the groundbreaking potential of 4D piezoelectric printing across diverse fields. In 2019, H. Cui *et al.* reported that with the design flexibility of additive manufacturing, it is now possible to manufacture complex architecture of piezoelectric materials that can manipulate electric displacement to selectively suppress, reverse, or enhance applied stress, opening doors for the next generation of intelligent infrastructure for automated bridges, actuators, and smart homes [2]. In 2021, Y. Wang and X. Li combined 3D printing and piezoelectric materials to propose a low-cost self-sensing intelligent actuator that can be used for multiple applications such as intelligent mechanical claws [1].

In the field of sensors, 4D printing of piezoelectric material has enabled the development of self-powered pressure and tactile sensors, with potential applications as internal organ data-receiving implants [6]. The application of 4D piezoelectric materials holds significant promise in medical domains, particularly bone tissue engineering, where their integration is pivotal. This significance is heightened by the well-established recognition of piezoelectricity's role in natural bones, a vital driver of successful bone regeneration. Utilizing 4D printing to create dynamic bio-piezoelectric scaffolds introduces programmable tissue electromechanical responses, a temporal dimension that responds to external cues, amplifying their potential for effective bone regeneration strategies. A. Chen *et al.* showed how the transformative potential of shape-shifting, functionally adaptable 4D bio-piezoelectric scaffolds holds promise for revolutionizing bone tissue engineering by enabling dynamic and personalized approaches to regeneration [5].

Current and Future Challenges

The rapidly evolving area of 4D printing of piezoelectric materials is characterized by a blend of present challenges and future possibilities that both shape and stimulate the field's advancement. In the present context, one of the foremost challenges lies in achieving complex 3D-printed structures of piezoelectric components. Achieving this requires unprecedented precision in managing material properties, meticulous control over fabrication processes, and a seamless alignment with existing technologies. Numerous reports have surfaced detailing the successful integration of various piezoelectric materials into intricate sensing mechanisms through 3D printing [2],[6]–[8]. In addition, several comprehensive reviews have highlighted the strides made in the realm of 3D printing for piezoelectric ceramic structures [9]–[11].

However, many challenges still exist to enable this technology into practical applications. Namely almost all piezoelectric materials possess inherently brittle characteristics, and a majority of them are ceramic compositions with high melting points, rendering direct 3D printing unviable using existing methods [10]. The current strategy involves amalgamating these piezoelectric materials with a polymer matrix to achieve printability, followed by the subsequent removal of the polymer via controlled polymer burning out after the completion of the printing process. One of the significant challenges in these parts is the low density and geometry distortion of the printed piezoelectric material [12], which determines the final piezoelectric properties of the structure.

The dynamic nature of these materials, to change and respond to external stimuli over time, ushers in another pressing challenge. Precisely modeling and anticipating their intricate reactions across different scenarios requires sophisticated computational methods and thorough experimental validation. There have been reports on 3D-printed piezoelectric actuators and intelligent structures utilizing the current computational capacity [13],[14]. However, it is evident that there exists substantial room for improvement within this domain.

Implants and bone tissue engineering stand out as significant applications for the 4D printing of piezoelectric materials. Nevertheless, specific challenges persist, such as: integrating multifunctional features for complex tissue regeneration needs, achieving precise stress-polarization conversion suitable for various bone tissues, refining non-invasive electromagnetic induction techniques for communication, and harmonizing multi-cellular behavior modulation across distinct scaffold microregions [5].

Aircraft, particularly intelligent morphing wings, stand as another promising domain for the utilization of 4D printing with piezoelectric materials. The ongoing experimentation with piezoelectric wings in aviation demonstrates the potential [15], and the design flexibility offered by 4D printing holds the capability to reshape the trajectory of aircraft and spacecraft industries. This could enable precise manipulation of wing motions during flights, contributing to a revolutionary shift in their performance and efficiency.

Another obstacle in applying 3D printing of piezoelectrics to create complex structures is poling and characterizing the complex part. Conventional poling systems, such as contact/thermal poling, fall short on non-flat complicated surfaces [10]. Although reports of in-layer in-situ poling techniques exist

[8],[16], their application restricts the height of final prints. Meanwhile, Corona poling, though not as effective, remains the sole option for now with prospects for future refinement. As for the characterization, the lack of standardized methods hinders systematic and exhaustive characterization of the printed 3D structures, hampering comparative analysis across diverse fabrication techniques [10]. Despite the potential 4D printing process for crafting intricate and adaptable structures, expanding these capabilities to larger scales while preserving precision and maintaining cost-effectiveness remains a significant obstacle, demanding innovative strategies that harmonize intricacy with efficiency, all while upholding affordability and practicality.

Advances in Science and Technology to Meet Challenges

Advancements in science and technology have been instrumental in driving innovation and addressing these obstacles posed by the 4D printing of piezoelectric materials. Researchers and engineers are diligently exploring diverse pathways to overcome the constraints and capitalize on the potential of this transformative technology. Within the domain of materials science, a focused endeavour is underway to convert brittle materials with enhanced flexibility and robust mechanical properties. These materials aim to mitigate the inherent brittleness of traditional ceramics, thereby facilitating their seamless and precise 3D printing [17]. Efforts are also directed toward the development of future 3D printing methods that enable the direct printing of ceramics [18].

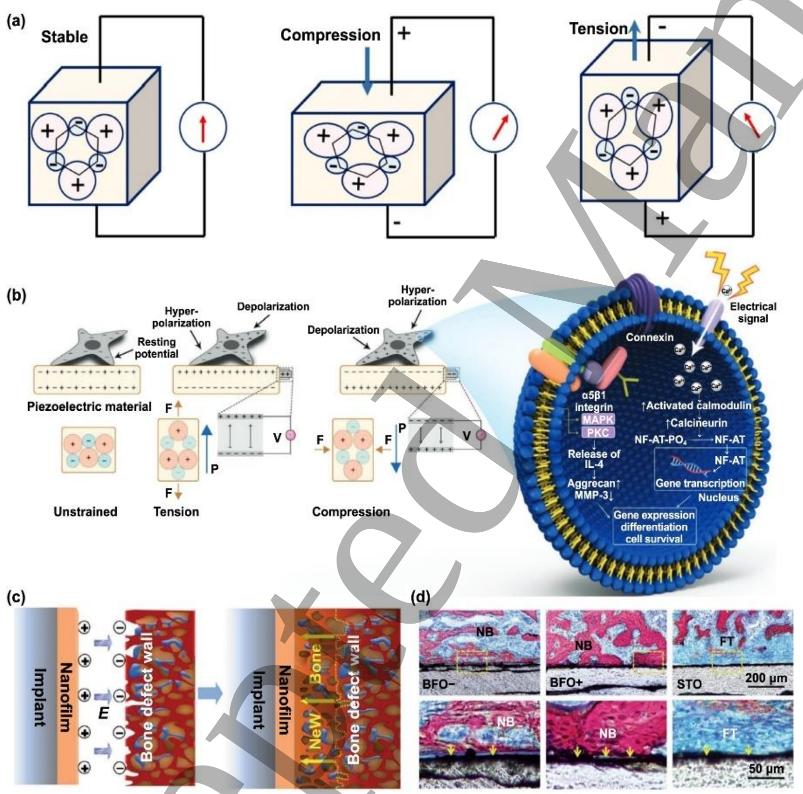

Figure 2: Schematic representation of (a) piezoelectricity of piezoelectric materials, (b) the stress-induced surface charges triggering cell signaling pathways, (c) Schematic illustration of the built-in electric field between the implant and native bone wall for osseointegration promotion, and (d) histological evidence for significant promotion of new bone formation using electropositive implant compared with electronegative and uncharged implant (Chen et al., 2023). Reproduced with permission from John Wiley & Sons. © 2017 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

Advanced simulations and modelling techniques are being developed to tackle the challenge of modelling and predicting the behavior of piezoelectric materials [19]. This software aids in understanding how these materials respond to different stimuli and facilitating the design of complex structures with predictable outcomes for different applications, such as aircraft wing morphing. The biotechnology and implant sectors are witnessing a growing emphasis on advancing 4D printing of bio-piezoelectric materials. The pursuit involves multimaterial 4D printing, coupled with inventive sintering approaches, aimed at creating precisely dimensioned multifunctional bio-piezoelectric implants [5]. The quest also extends to devising non-invasive and secure communication mediums. However, ensuring the consistency of 4D-printed structures is of utmost importance, enabling their seamless integration into practical applications with confidence. Figure 2 illustrates the schematic depiction of the operational principle of piezoelectricity, stress-induced signal charges, and the potential of piezoelectric implants in facilitating bone formation. The biotechnology and implant sectors are witnessing a growing emphasis on advancing 4D printing of bio-piezoelectric materials. The pursuit involves multimaterial 4D printing, coupled with inventive sintering approaches, aimed at creating precisely dimensioned multifunctional bio-piezoelectric implants [5]. The quest also extends to devising non-invasive and secure communication mediums. However, ensuring the consistency of 4D-printed structures is of utmost importance, enabling their seamless integration into practical applications with confidence. Figure 2 illustrates the schematic depiction of the operational principle of piezoelectricity, stress-induced signal charges, and the potential of piezoelectric implants in facilitating bone formation.

Additionally, collaborative efforts across disciplines are fostering creative solutions to address challenges such as stress-polarization conversion and the modulation of multi-cellular behavior [5]. Teams of engineers, biologists, and material scientists are working in unison to design implants with specialized functions.

Furthermore, there is an active pursuit of advancing 4D printing technologies to improve scalability, cost-effectiveness, and precision. For large-scale printing, new printing techniques are being explored to enable the creation of larger and more complex structures while maintaining high accuracy [20]. These advancements are vital for enlarging the scope of 4D-printed piezoelectric materials to practical scenarios.

Concluding Remarks

Advances in material science, computational modeling, additive manufacturing, and biotechnology are collectively shaping the trajectory of 4D printing of piezoelectric materials. This revolutionary technology, propelled by cross-disciplinary collaboration and cutting-edge advancements, promises to reshape diverse sectors. While challenges such as the natural property of brittleness, poling strategy of complex structure, and precise modeling persist, they serve as catalysts for innovation. Through addressing current challenges and pushing the boundaries of technological feasibility, these advancements are opening up novel possibilities for the integration of piezoelectric materials across a broad spectrum of applications, from medical implants to smart structures and beyond.

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9 – 4D Printing in Soft Robotics

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Status

In recent years, the field of soft robotics as seen the introduction of 4D printing technology. Soft robotics is a specific branch within the subject of robotics that concentrates on the conceptualization and advancement of robotic systems using flexible and deformable materials [1]. The historical origins of the integration of soft robotics with 3D printing technology may be traced back to the year 2009 [2], although many studies have shown the use of 3D printing in biomimetic applications for the last two decades [3]. Numerous factors must be considered to successfully integrate 4D printing into the field of soft robotics. Unique design methodologies, additive manufacturing procedures, the use of smart materials in the printing of soft devices, geometrical optimization, and the control techniques used for properly operating soft robots are some of the key components of this discipline. Each of these facets has been mentioned by researchers recently to pave the way for faster advancements in soft robotics field on a more reliable and sustainable path.

Within the classification framework established by the American Society for Testing and Materials (ASTM), there are seven distinct categories denoted as 7 additive manufacturing techniques. Notably, four of these categories, namely material extrusion, vat photopolymerization, material jetting, and powder bed fusion, find use in the production process of 4D-printed smart materials in robots manufacturing. For programmable 4D-printed objects, design necessitates computational modelling and topological optimization. In this regard, Zolfagharian et al. [4] recently developed a comprehensive finite element model to determine the optimal shape and thickness of a soft gripper blade to ensure that it could grip objects of assorted size and shapes. Also, researchers have recently used the machine learning technique as a flexible and effective tool to anticipate the actuation mechanism of 4D-printed soft robotics. Hereof, Sun et al. [5] proposed a novel machine learning- and evolutionary algorithm-based technique for designing a 4D-printed active soft actuator. Moreover, machine learning is another contemporary technology that has gained more attention in smart material design. In general, the integration of machine learning in the design process, together with the utilization of 4D printing, presents a promising methodology for forecasting desired characteristics, implementing reverse engineering, and simulating intricate shapes of smart materials used in the field of soft robotics [6].

To comprehensively examine all the variables involved in the 4D printing of soft robotics, it is essential to carefully evaluate and implement suitable control strategies. Due to the nonlinear nature of soft robotics, several innovative control techniques have been developed. Mohammadi et al. [7] discussed one of the most common techniques for autonomous controls using reinforcement learning technology to control a 4D-printed variable stiffness structure in soft robotics. **Figure 1** presents a concise overview of the sequential steps included in the manufacturing process of 4D-printed soft robotics.

Current and Future Challenges

At present, the utilization of 4D printed soft robotics is primarily observed in laboratories, prototyping facilities, select manufacturing exhibitions, and a few medical apparatuses. As depicted in **Figure 1**, the initial phase in the manufacturing process of 4D soft robotics is the design and optimization of preprogrammed smart materials. One of the primary obstacles encountered in this field is to the inherent complexity associated in modelling and forecasting the movements of actuators and soft robots, mostly due to the nonlinear characteristics shown by the materials involved. Finite element analysis has been shown to possess many drawbacks. One instance of mistake arises from geometric approximation, whereas the finite element analysis approach is characterized by its computationally intensive nature, mostly attributed to the substantial number of degrees of freedom involved. However, machine learning-based methodologies have the potential to address these issues [8].

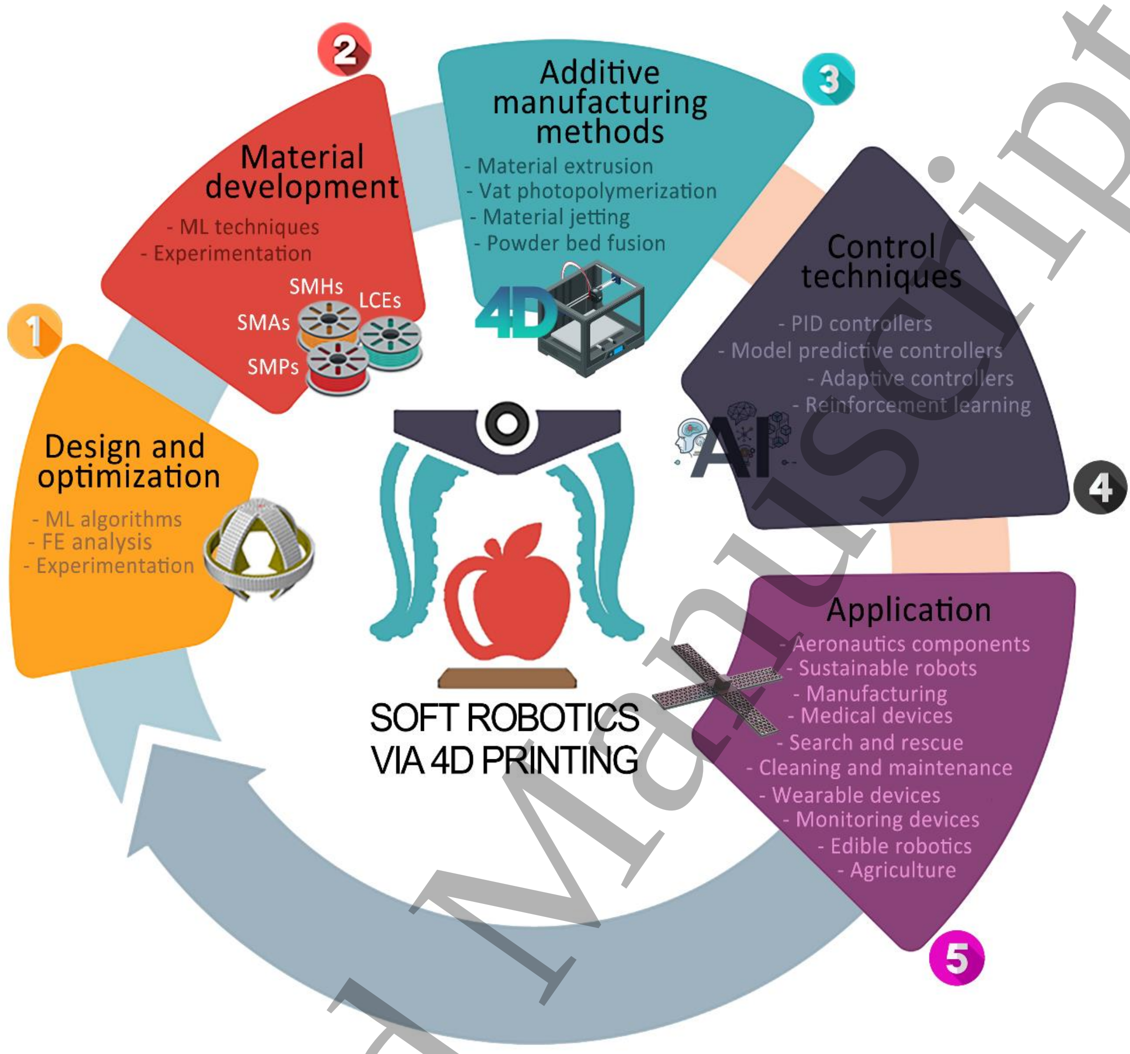

Figure 1. Sequential steps included in the manufacturing process of 4D-printed SR: 1-design and optimization, 2-material development, 3-additive manufacturing methods, 4-control techniques, 5-proposed application of designed SR.

Furthermore, the range of available materials for 4D printing of soft robotics is limited. This issue presents a significant challenge as not all polymers possess the requisite properties to undergo shape transformation. The aforementioned characteristics encompass rigidity and stiffness, surface smoothness, dimensional stability, degradation, and the formation of voids [9]. Recent studies also have addressed the high demand for a sustainable world that employs biodegradable materials in the near future. Therefore, the development of novel smart materials with biodegradability characterization will pose a significant challenge in the field of 4D printing of soft robotics soon [10].

At present, most 3D printing machines exhibit a closed architecture, thereby limiting the ability of researchers to make substantial modifications to processing conditions. The 3D printers are engineered with a fixed process parameter that necessitates a trade-off between the precision of the printed parts and the speed of the printing process. One of the primary obstacles in the advancement of 4D printing of soft robotics is the challenge of attaining real-time dynamic control over printing parameters. This challenge requires the synchronized enhancement of both the hardware and software components of 4D printing technology [11]. The deformation process of soft robotics components is characterized by its simplicity, yet there lacks a well-established theoretical framework that possesses the necessary level of advancement to accurately forecast the resultant shape of the soft robotics components following deformation. It is imperative to thoroughly consider the micro-composition of the material, the shape-memory mechanism, the manufacturing process, and the process parameters. In addition, it is imperative to develop novel theories and simulation techniques [12].

In soft robots, soft electronics, and medical devices, the components produced through 4D printing are required to exhibit prompt responsiveness to environmental stimuli. This necessitates the achievement of rapid reaction times and precise control over the system. Achieving rapid response and precise regulation solely through the manipulation of ambient temperature, humidity, or electric heating poses a difficult challenge. Consequently, it is vitally important to conduct research on emerging smart materials, control methodologies, and application contexts pertaining to 4D printing technologies [13].

Advances in Science and Technology to Meet Challenges

Deep learning, artificial intelligence, and machine learning (ML) are widely used in the field of smart materials and soft robotics design in the present day. The ML model learns from the input data sets that are used to predict future outputs. It is a versatile and effective tool for predicting the actuation mechanism of 4D-printed soft robots during the design phase, which could save time and cost and enhance precision [14]. In this regard, Zolfagharian et al. [15] conducted a study on ML and finite element-based models to accurately predict the specific bending angle of a 4D-printed soft pneumatic actuator. It was shown that ML models possess significant potential for optimizing hyperparameters and have 94.3% accuracy. Furthermore, as previously stated, ML-based algorithms are utilized to fulfill the requirement for innovative design of novel smart materials in the field of 4D printing technology. Dutta et al. [16] have proposed a ML-assisted approach for the development of novel shape memory polymers using polyurethane films. The proposed methodology offers a rapid means of characterizing newly discovered materials by integrating experimental findings with predictive modeling techniques. The modeling workflow that has been presented demonstrates a predictive accuracy of 90% in forecasting the recovery behavior of polyurethane films. Considering the various control algorithms, ML has also become an applicable instrument. Mohammadi et al. [7] 4D printed a variable stiffness joint based on shape memory material with polylactic acid filament, as shown in **Figure 2.** Based on the reward function optimization concept, the system was controlled using the reinforcement learning (RL) approach. The results demonstrated a great potential for the RL controller to adjust online to numerous unanticipated circumstances.

The utilization of multi-material printing has already facilitated the fabrication of intricate structures possessing diverse properties [17]. Furthermore, the advancement of multi-material 4D printing methodologies, which ease the incorporation of multiple stimuli-responsive materials within a singular structure, has the potential to expand the range of potential applications [18]. In addition, researchers have successfully incorporated continuous fiber-reinforced in 4D printing with soft robotics to respond to the requirements of advanced smart materials that possess enhanced mechanical properties. Wang et al. came up with a 4D printing method for liquid crystal elastomers that uses continuous fiber reinforcement to make soft robotics arms that can bend and stand on their own. The research demonstrates that the printed composite can carry a load of up to 2805 times its own weight and achieve a bending deformation curvature of 0.33 mm^{-1} at 150 °C.

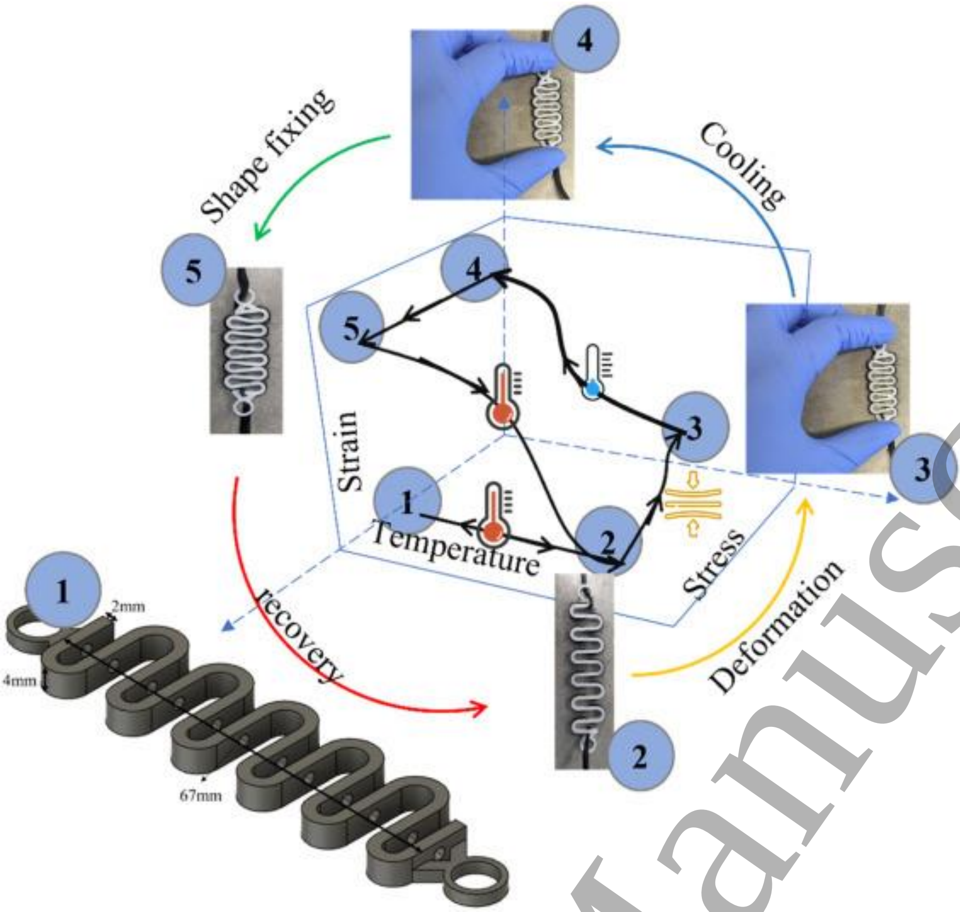

Figure 2. 3D-printed spring designed to provide variable stiffness joint in flexible robots [7].

Concluding Remarks

The utilization of 4D-printed soft robotics has the potential to significantly impact the application possibilities of robots across various domains. The utilization of 4D printing technology in the process of soft robotics fabrication has the potential to contribute to the realization of a more sustainable global environment. By directing research efforts towards this area, it is anticipated that new possibilities within the field of soft robotics will be discovered in the coming years. Significant effort is necessary for the advancement of soft robotics applications in the foreseeable future, to establish 4D printing as a widely adopted technology, comparable to 3D printing technology. This can be achieved through the integration of innovative design methodologies utilizing artificial intelligence capabilities, the exploration of diverse and readily accessible smart materials suitable for 4D printing technologies, and the utilization of micro-fabrication techniques with adjustable parameters. The exploration of new control techniques and application scenarios is of utmost importance in the context of 4D printing components. These components are required to respond promptly to environmental cues, which in turn demands quick reaction times and precise control. Moreover, physics-informed machine learning serves as a highly efficient, robust, and adaptable instrument, thereby constituting an additional area of investigation within the research community focused on 4D-printed soft robotics.

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10– 4D Printing of Dielectric Elastomer Actuators

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Status

4D printing of dielectric actuators (DEAs) combines multi-material 3D printing technologies with conductive and dielectric polymers and polymer composites. It enables an automatic single-process fabrication of individualized actuators [1]–[3], which deform based on the applied voltage to the electrodes. Historically, DEAs were broadly researched for their advantages of light-weight solid-state design, fast response time, and silent operation, to name a few, but were limited by cumbersome manual fabrication techniques, which limited their utilization [4].

Recently, 3D-printed DEAs, utilizing extrusion-based 3D printing like fused-filament fabrication (FFF) [1] and direct ink writing (DIW) [2], showed that 4D printing can provide a necessary catalyst to make DEAs more readily available. In addition, 4D printed actuators can be combined with 3D-printed sensors [5], [6] and conductors [7] into seamless 4D-printed multi-functional smart structures, which cannot be achieved with conventional technology.

The field of 4D-printed DEAs is being propelled forward by novel technological and material developments. On the materials front, novel conductive polymer nanocomposites and engineered self-healing dielectrics with high permittivity and flexibility are increasing the efficiency and applicability of 4D printed DEAs [8]. On the technological front, the 3D printing process is supplemented with additional processes to achieve the desired material and surface properties, i.e. in-situ polymerization of the dielectric [2], [3] and ironing of each dielectric layer [1]. These developments show promise in broadening the applicability of 4D printed DEAs in soft robotics, human-machine interaction, medicine, vibroacoustic, and optical applications [9].

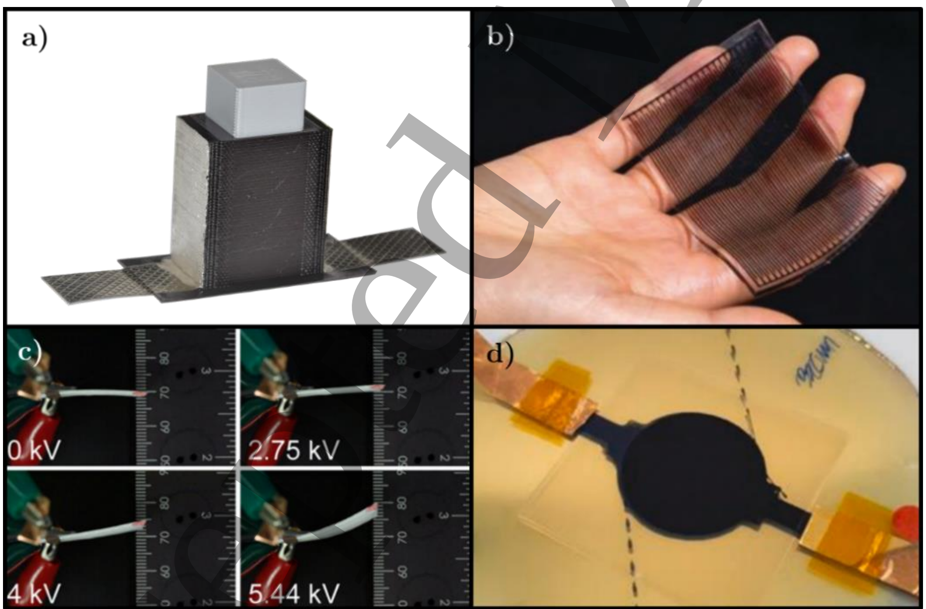

Figure 1. Examples of 3D printed DEAs: a) dynamic stacked DEA [1], b) interdigitated DEA with self-healing capability [3], c) bending DEA, 3D printed with Direct Ink Writing [2], d) planar DEA for optical applications [20].

Current and Future Challenges

4D printing of DEAs uncovers several technical limitations and issues that need to be addressed. A primary technical challenge lies in the printing accuracy: the performance of 4D printed dielectric structures is constrained by the thickness of the dielectric and conductive layers [10], [11]. Current 3D

printing techniques based on material extrusion are limited by the minimum layer thickness, which can limit the performance of the final product. Moreover, achieving flatness, homogeneity, and minimal porosity in these layers is challenging [1], [11]. Imperfections, such as uneven surfaces or internal voids, can lead to dielectric breakdowns and introduce instability in the printed structures [11], [12].
Material limitations present the next challenge in 3D printing of DEAs. From a mechanical standpoint, the stiffness of the material determines the achievable deflection, while its damping properties can lead to issues like heating and energy loss [10]. On the functional side, the conductivity of the electrodes and the permittivity of the dielectric, influenced by both material selection and filler content, are critical for the performance of DEAs [2], [3], [10]. The material properties like stiffness, damping, and filler content play a crucial role in the 3D printing process, introducing challenges related to printability [13]. Therefore, there's often a compromise to be made between mechanical and functional properties and printability of the selected or developed material.
Reliability, especially in 4D-printed DEAs, remains an area requiring more extensive research. While conventional DEAs have demonstrated good reliability over extended operation [14], [15], the same hasn't been extensively validated for 3D printed DEAs.

Advances in Science and Technology to Meet Challenges

The future of 4D printing hinges on the evolution of both 3D printing technologies and materials. For the technology, in-process monitoring, and closed-loop control will be crucial for precise, consistent, and repeatable outputs [16]. Moreover, 4D printing will significantly benefit from integrating multiple technologies, such as combining FFF, DIW, Inkjet, and incorporating additional processes like in-situ polymerization [17].
On the materials front, there's a pressing need to enhance the properties of 3D printing materials. This includes increasing the conductivity, permittivity, and printability of polymer composites, and enhancing their flexibility [10].
Driving these technological and material advancements are applications demanding individualization. 4D printing can revolutionize the fields of individualized prosthetics and haptic feedback systems [18], while industrial applications, particularly in milli-, micro-, and nano-positioning systems, and vibroacoustics [19], such as active noise isolation, are also on the horizon.
Recently, there's been a surge in investment and commercial development in the industrialized manufacturing of stacked DEAs, highlighting the industry's recognition of the unique capabilities these actuators offer.

Concluding Remarks

4D printing of dielectric actuators offers a streamlined, single-process fabrication method that overcomes the limitations of traditional manufacturing techniques. Recent advancements in materials and technology are expanding the applicability of 4D-printed DEAs across various fields, from medicine to industrial applications. The ongoing research and investment in this area signals its growing importance and potential for widespread adoption.

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11 – 4D printing of Bio-inspired Musculoskeletal Systems

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Status

An artificial musculoskeletal (MS) system represents a bio-inspired structure, leveraging a unique blend of soft, semi-soft and stiff materials to emulate biological systems that underpin the movement and functionality of animals [1-3]. This structure fills the gap between entirely soft and rigid robotic structures, offering a harmonious blend of precision, high load carrying capacity and energy efficiency. Applications for this technology span a wide spectrum, ranging from bio-inspired prosthetic devices to robotic arms, endoscopes, biomimetic robots and humanoids. These systems consist of intricate geometry and encompass various structures resembling bones, muscles, tendons, ligaments, nerves, and sensors. Manufacturing of MS systems can be categorized as "multimaterial 4D printing with soft, stiff and active materials", which consists of various novel aspects.

Multimaterial 4D printing is relatively new compared to the history of 4D printing, which was conceived in 2013 by Tibbits. The core of this technology lies in the seamless integration of active and passive materials within a single framework, demanding an advanced additive manufacturing system and an intelligent mechatronic system design. Active materials, or smart materials in this context, are those responsive to external stimuli such as conducting polymers (CP), dielectric elastomers (DE), shape memory polymers (SMP), piezoelectric (PZ), shape memory alloys (SMA), and polymer gel actuators (PG) and Twisted and coiled Polymer (TCP) muscles. In contrast, passive materials include those structural materials used in various additive manufacturing systems.

The landscape of multimaterial 4D printing has been marked by major development across various scales and domains, in microdomain and nanodomain manufacturing. Smart materials, especially shape memory polymers have been extensively shown for 4D printing [4-6]. While considerable effort has been made to manufacture MS system using 3D printing setup, significant challenges persist. A recent review paper showed the effort on multimaterial 4D additive manufacturing addressing key challenges [4, 7]. The major breakthroughs and advancements in this field are related to: i) *improved printing accuracy and speed*, ii) *seamless integration of different materials within a single robotic structure* [7, 8], iii) *multifunctionality incorporating sensory capabilities and even energy harvesting, all integrated into a single robotic system* [9], iv) *biobybrid systems that use living cells in synthetic materials in a multimaterial 4D printing* [10].

A schematic diagram and some attempts of manufacturing of a musculoskeletal system are shown in Fig.1, emphasizing on multimaterial with active materials that serve as actuators, other passive materials with nanomaterials to mimic bone-like structures, and soft materials to replicate biological tissues. In this effort, our group presented various MS systems using artificial muscles, using fiber reinforced PLA and silicone [11], and a joint mechanism using novel TCP artificial muscles [12]. Recently, we showed a multimaterial one shot manufacturing, using both active and passive materials to fabricate a robotic finger in one build [13]. While a significant advancement has been made in additive manufacturing of multi-material, through the polyjet technology and others, limitations exist on printing active materials and concerns about their reliability. Nevertheless, the pursuit of creating ball and socket cascaded structures remains a promising frontier [14]. This development will advance the creation of advanced articulated joints that mimic nature with all the benefits such as agility, flexibility, and energy efficiency. This may enable us to realize the entire humanoid robot additively manufactured using 4D concepts.

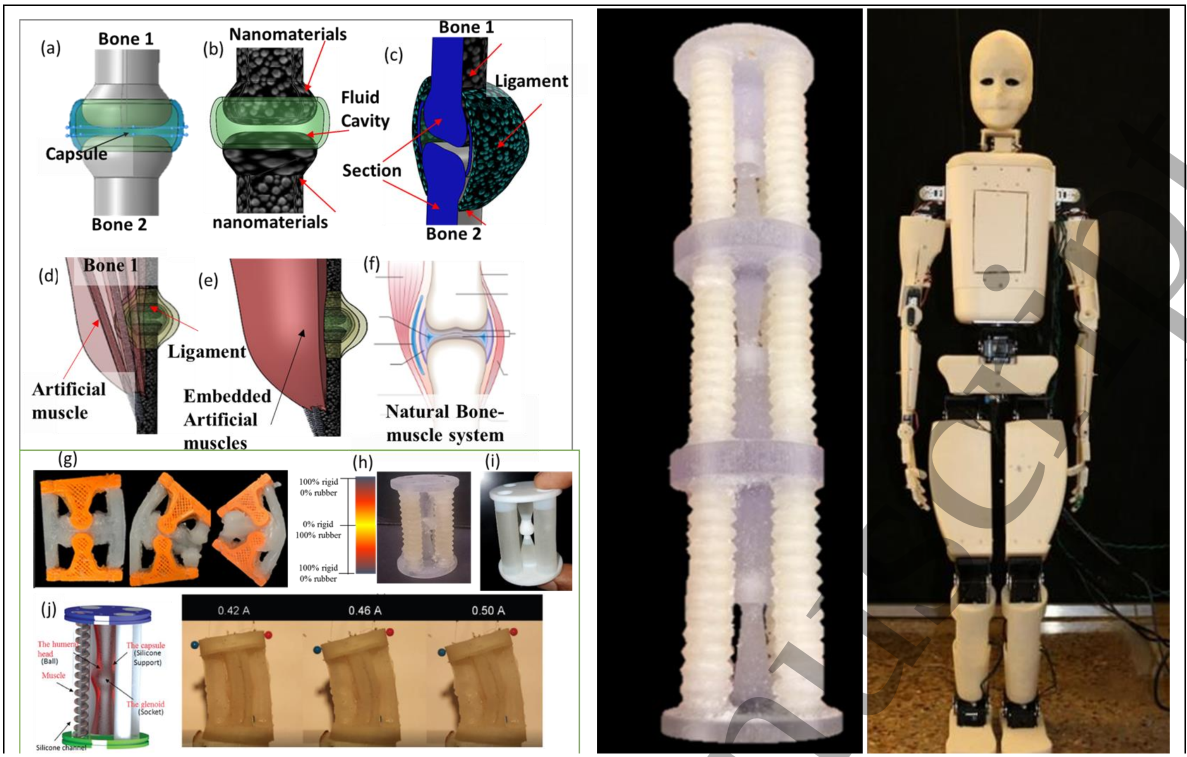

Figure 1. Bioinspired musculoskeletal system additive manufacturing **(a)-(f)** conceptual realization of synthetic material mimicking muscles, bones and soft tissues; **(g)** Metal fiber reinforced PLA and soft silicone, 110 mm in height (Adapted from Elsevier) [11], **(h)** & **(i)** functionally graded material (FGM) structure with soft/stiff structure using polyjet technology (Adapted from Springer) [14], **(j)** MS system with TCP muscles via molding (Adapted from Wiley) [12]. **(k)** cascaded soft and stiff materials using Connex printer and **(l)** humanoid robot 120cm in height hat has been developed at the HBS lab for potentially 4D printing MS system. These prototypes are fabricated using custom made setup and advanced 3D printers.

Current and Future Challenges

There are several challenges in making muscle systems in additive manufacturing due to the limited knowledge on processing parameters, materials suitability, interface structure, control aspect, and performance of the product such as reliability. To realize 4D printing, fluidic elastomer actuator units [8] have been used in many demonstrations. However, pneumatic systems have their own disadvantages, such as acoustic noise, the need for bulky compressors and portability issues. Biomanufacturing using inks is another approach using living cells and scaffolds. However, there are many complicated issues related to materials, processing steps and technologies employed. In addition, there has not been a commercialized 3D printer specifically for 4D printing and hence limitation [4].

To realize a musculoskeletal system without dealing will living cells, key knowledge on the fabrication of functional systems utilizing active and passive materials is required. The key point is that a good understanding of structure-property relationships during systematic manufacturing or co-fabrication of active and passive materials is needed to fully realize functional dynamic systems. This approach could help overcome many of the challenges in making new device and systems. Currently, there is limited knowledge as to how materials behave during co-fabrication, particularly in the artificial muscles and stiff structures arena. In the domain of soft robotics, stiff structures connected by soft joints are highly appealing because of their smooth operation and safety [15]. Researchers have focused on utilizing additive manufacturing technology for both stiff and soft materials [16-18]. One of the major advancements recently was a hybrid manufacturing approach of active biorobotic structures shown in Fig. 2 that incorporates technology from the fused filament fabrication (FFF) process, direct-ink-writing (DIW) process, and "stop and go" manufacturing process using a pick and place robot (PPR) [13]. The challenge for this is the software interface and the arrangement of multiple muscles that need further research.

Various robots were designed and fabricated using 3D/ 4D printing approach recently. In some cases, the actuators were integrated after the parts were manufactured. In other cases, the active materials are printed according to the design. If all the parts are printed all together (the actuators, the batteries, and all the accessories) following 4D printing principles, a fully functional device or system will be obtained directly from the manufacturing setup. To accelerate the fabrication of such technology, high energy density artificial muscle manufacturing in additive manufacturing systems is extremely important. Researchers have printed shape memory polymers via 3D printing; however, their force output is small compared to shape memory alloys (SMA), hence, high energy density materials as active materials along with modelling and simulations should be explored. In addition, existing multimaterial 3D printers have issues on reliability, elasticity and they are expensive too. Hence, new processes and custom-made setups are needed.

Advances in Science and Technology to Meet Challenges

4D printing of multimaterial will have significant impact in the healthcare sector as it enables us to create functional dynamic structures. This will bring a new opportunity on a custom-made setups or new research methods that enable us to realize a multimaterial manufacturing process that includes both stiff and soft composite material as well as actuation units, making a functional device from a computer aided design (CAD) model in a single manufacturing batch. New manufacturing setup should be explored for industrial applications and commercialization for making robotic systems directly from CAD files including components such as batteries and electrical wires. The future research objectives should address critical aspects to contribute processing parameter knowledge and help in the invention of a machine that makes all the necessary features to create functional devices and systems. We may see such process and machine soon in the next few years, However, there is a lack of information on the behavior of materials, the processing conditions, and structure-property relationships of co-fabrication of active and passive materials in AM systems. Another issue is that multimaterial 4D printing may introduce new regulatory and safety challenges, particularly when used in applications like healthcare or aerospace, where strict standards and certifications are required. The expectation is that there will be more materials that can be co-fabricated, new processes and custom-made setups and new algorithms to coordinate different technologies to fabricate bioinspired robotic systems. These are new aspects that should be addressed in future research. This could be done either by customizing current approach and setups or combining different technologies.

Based on the current trend and the need for new directions to meet the challenges, the future research should focus on:

1) *Modeling and Simulation*: Modelling and simulation to investigate the correlation of material composition, processing parameters, and the resulting fabricated structures.
2) *Customized Setups:* Customized manufacturing setups that allow studying extremely soft (modulus 8 kPa @100% strain and maximum elongation an order of 1200%) [19, 20] and high energy density materials (10^5-10^7 J/m^3).
3) *Employing Computer Vison*: Vision assisted manufacturing to automate the placement or embedding process of active materials.
4) *Machine Learning and Artificial Intelligence*: Incorporating machine learning in AM to predict the behavior of fabricated structure from imaging data and other data sets.
5) *Novel Approach*: New process that shows unique 4D structure/ machines, which could be adapted from how nature creates such elegant musculoskeletal system that grows in time.

This may require collaboration between academia and industry to push the challenge in terms of bringing the technology to end users and solving the fundamental challenges. Establishing intellectual property rights and industry standards for multimaterial 4D printing is crucial for ensuring fair competition and interoperability.

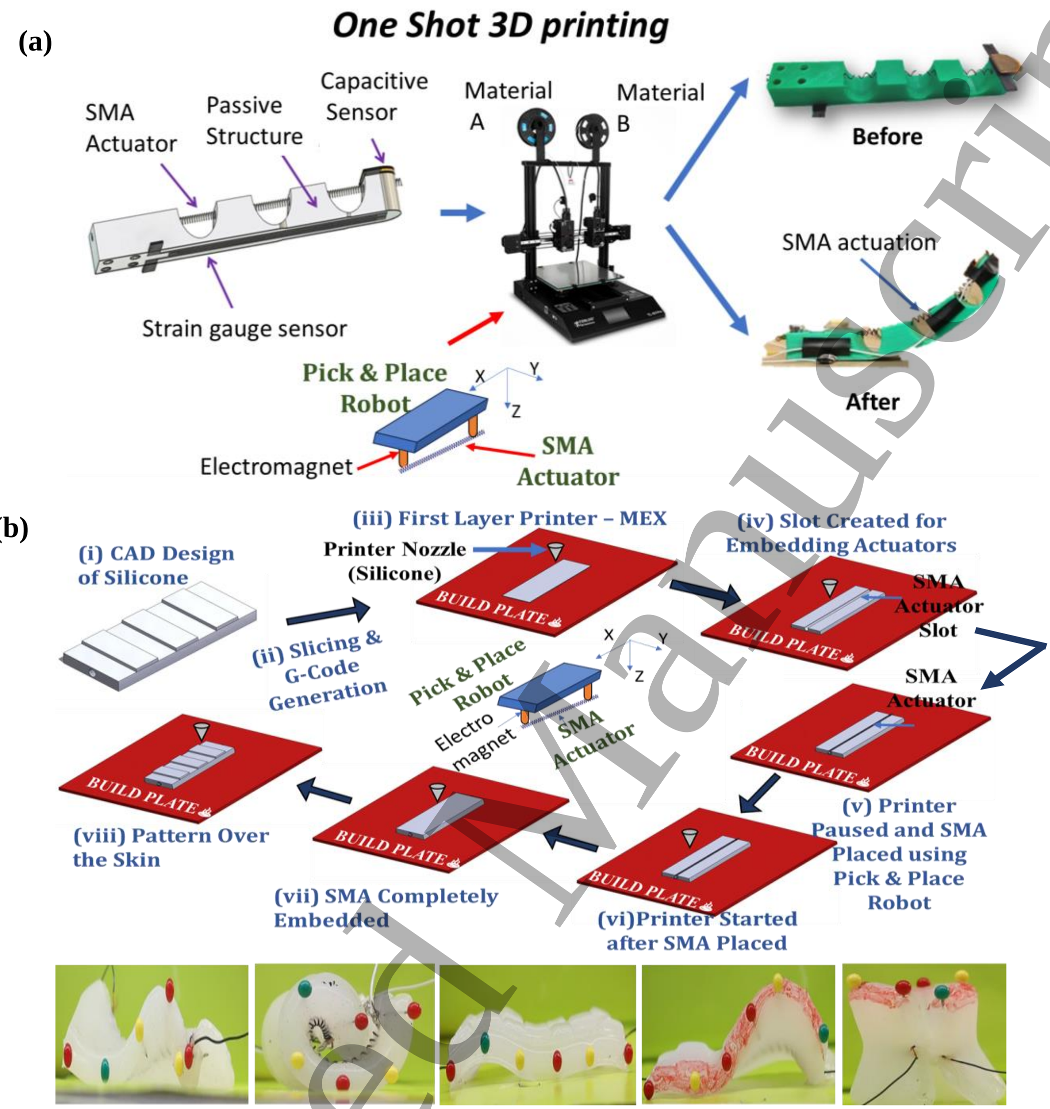

Figure 2. One shot manufacturing approach of robotic structures: Direct ink writing (DIW), material extrusion (MEX) and Pick and Place robot (PPR) based additive manufacturing: **(a)** manufacturing of morphing structures actuated by SMA springs and soft silicone along with pattered 3D printed silicone (Adapted from IOP publication) [19] and **(b)** one short 3D printing of robotic finger along with sensors and SMA actuators (Adapted from Springer) [13].

Concluding Remarks

An artificial musculoskeletal (MS) system is a typical bioinspired multimaterial system that can be realized with the concept of 4D printing. Such structures need a new manufacturing method, which should enable the fabrication of artificial bioinspired systems comprising active materials (artificial muscles and sensors) and passive materials (tendons, ligaments, wiring system). This can improve the way we manufacture devices in a fully automated manner in 4D concept. The approach proposed for fabricating musculoskeletal systems using nanomaterials infused thermoplastic materials for stronger structures, employing novel interface at the stiff/soft joint, embedding, or creating actuation unit, and employing bio-inspired joints in the modular system, can be easily extended to manufacture other devices needing both active and passive materials. By way of such extension of 4D printing of multi materials, we can ultimately make an artificial finger/hand or hand orthosis that has flexible and stronger joints. The process will be extremely useful for manufacturing soft robots with continuum structures, even manufacturing the entire humanoid robot for future home assistant or co-robot. In addition to the societal benefits in healthcare, automated manufacturing techniques in artificial MS system offers insights into smart materials, additive manufacturing, and biological systems, enabling a deeper understanding of these domains.

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12- AI-based Control in 4D Printing

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Status

Advancements in various disciplines, such as material science and manufacturing engineering have promoted the emergence of 4D printing as a new fabrication technique for producing complex and versatile robots. From a control engineering perspective, the absence of models and controllers for the 4D-printed robots in the current studies have hindered the wider applications of these new robots in industrial scenarios. Currently, a dedicated body of research is directed towards refining the modeling and control aspects of 4D printing [1,2]. These investigations collectively validate the enhanced performance exhibited by 4D-printed robots. Notably, a significant proportion of these studies adhere to conventional methodologies within the domain of control engineering. However, considering the rapidly escalating capability and complexity characterizing 4D-printed robots, the demand for more sophisticated tools becomes imperative. In this context, AI-based techniques emerge as highly promising solutions.

AI technologies can be generally characterized into supervised learning, unsupervised learning, and reinforcement learning. Each of these learning paradigms offers unique advantages that can be leveraged to enhance the control of 4D-printed robots. Supervised learning can be utilized to train models using labeled data, enabling accurate predictions of the robot performance given a stimulus. Unsupervised learning, on the other hand, can help discover hidden patterns in unlabeled data. However, the related applications in 4D-printed robots are limited. Lastly, reinforcement learning can enable robots to learn optimal policies through interaction with their environment, thereby promoting autonomous decision-making capabilities.

Current and Future Challenges

Due to the employment of soft and smart materials, the dynamics of 4D-printed robots are normally highly nonlinear and time-variant, which makes the modeling and control challenging [3]. Nonlinearity stems from the fact that the behavior of these materials is complexly linked to their physical properties, such as elasticity, damping, and their inherent properties like shape-memory behavior. The temporal variance of system dynamics is another significant challenge [4]. In a 4D-printed robot, the smart material's ability to change shapes or properties over time can introduce time-dependent variability into the system. This time variant nature of 4D printing introduces additional layers of complexity to the control problem. Another challenge is the morphing hysteresis inherent to many smart materials such as shape memory polymers [5]. Hysteresis is a time-dependent, nonlinear phenomenon where the next state is dependent on not only its current state and current input but also past states and/or past inputs. Lastly, the multi-domain nature of 4D-printed robots introduces further challenges [6]. Recent studies have the tendency to employ more advanced materials and more complex designs to achieve more functional robots, which makes the modelling and control even more challenging.

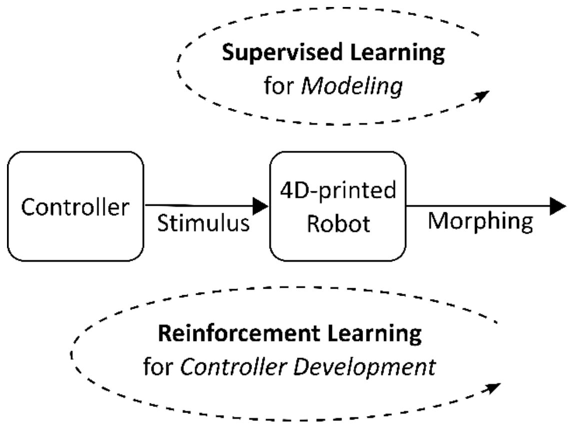

Figure 1. The diagram depicts how Artificial Intelligence (AI) technologies are used in 4D printing control. Supervised learning is used to create a model of the 4D-printed robot by studying the stimulus and the robot's morphing. Reinforcement learning is used to build the robot controller by interacting with the robot and observing its reactions.

Among AI-based methods, supervised learning and reinforcement learning stand out as the most influential tools for managing the aforementioned complexities as shown in Figure 1. Supervised learning, through the aggregation of a substantial volume of observational data pertaining to the robot's response to specific inputs, can directly fit a model. This process effectively bypasses the need for an in-depth analysis of robot physics. However, it's important to acknowledge that supervised learning is heavily reliant on both the volume and quality of data. Furthermore, the selection of model representations and the training process are largely dependent on human expertise.

On the other hand, reinforcement learning algorithms are exceptionally well-suited for executing closed-loop control tasks. By engaging with the 4D-printed robot, reinforcement learning can autonomously train a control policy that optimizes accumulated rewards. These rewards are structured in such a manner that greater rewards are granted when the robot's performance aligns with human expectations. An additional benefit is that reinforcement learning is capable of online learning. This means that the control policy can continue to be refined post-deployment, effectively managing the time-variant property of 4D-printed robots. However, it's worth noting that reinforcement learning also has its limitations, such as a tendency to become trapped in local optima and a necessity for meticulously designed reward functions. These characteristics underscore the need for algorithm developers to possess a high level of expertise.

Advances in Science and Technology to Meet Challenges

According to our investigation, most of the applications have been using AI to model the complex dynamics of 4D-printing, some using AI models as the controllers to control the robot in a closed loop. A few reports were found regarding parameter tuning.

One example that has been systematically elaborated is the control of 4D-printed Shape Memory Polymers (SMP). The use of traditional PID controllers can facilitate accurate deformations in SMP actuators, even in the presence of fabrication errors and actuation disturbances [7]. However, these controllers are static and struggle to manage the variable properties when the SMP samples undergo multiple deformations. Conversely, the incorporation of reinforcement learning in subsequent studies significantly improves control performance, achieving optimal results for given requirements [8]. To sustain control performance with time-varying properties, adaptive controllers have been further developed based on Q-learning. These controllers adjust the control parameters using the real-time observations of the SMP morphing and can maintain high precision control even when the shape memory effect is degraded [9].

The use of AI in modeling 4D printing is another rapidly evolving research area. Zolfagharian et al. [3] employed neural networks to model and predict the shape transformation of soft pneumatic actuators, achieving high precision and time-efficient predictions. Similarly, Yu et al. [10] leveraged the time-efficiency of machine learning models compared with numerical simulations, using a random forest for swift deformation predictions. Beyond modeling the 4D morphing process, AI models can also describe the fabrication process and predict how manufacturing parameters influence the performance of 4D-printed robots. For instance, Su et al. [11] compared various machine learning algorithms such as random forest and gradient boosting to model how printing parameters affect morphing behavior. Their results also highlighted which printing parameters have a greater impact on morphing properties. Sun et al. [12] integrated machine learning with an evolutionary algorithm to predict shape morphing for different distributions of active and passive components in printed samples. Their findings contribute to streamlining the transition from design to manufacturing.

Concluding Remarks

With the rapid development of 4D-printed robots and their increased complexity, AI-based methods serve as a very powerful tool for modeling and control. In the modeling domain, we consider supervised learning as a suitable tool to represent the complexities of 4D printing in terms of non-linearity and time-variance. With the development of large models in recent years, AI models will have stronger ability to tackle the large amount of data collected during the design, simulation, and manufacturing phase, which we foresee as a potential research area to enable a better prediction of the properties of the robots in an earlier phase. The studies on AI-based parameter tuning for developing controllers are currently limited but remains promising as it increases the automation and adaptation level, which fits well to solve the time-variance of 4D printing. Similarly, reinforcement learning can also generate control policies automatically while increasing the ability of controller models. This enables the formulation of optimal and adaptable control strategies, even in the absence of a priori knowledge about the robot's dynamics.

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13 - 4D Printing of Soft Sensors

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Status

In 2013, Tibbits introduced 4D printing, an innovative offshoot of 3D printing that combined 3D printing with time [1]. This innovative approach seamlessly integrates the principles of 3D printing with the dimension of time, effectively fusing physical form with temporal dynamics [2]. Originally focused on shape changes over time, the concept has now expanded and encompasses structures that alter shape, properties, and function in response to external factors like heat, water, light, and pH [3]. This approach has been harnessed for crafting functional devices like actuators, soft electronics, and soft sensors, offering novel opportunities across industries. Within this spectrum, sensors hold a pivotal role in the realm of wearable electronics, facilitating mechanical interactions between humans/machines and their environment [4]. They fall into several categories, such as piezoelectric, piezoresistive, capacitive, triboelectric, and magnetic—each with distinct mechanisms (5). Traditional soft sensor manufacturing involves intricate processes like film patterning, metallic deposition, and molding, which prove complex and costly. Alternatively, multi-material AM enables swift and cost-effective fabrication of soft sensor materials, endowing them with functionality, aligning with the concept of 4D printing. Up to now, many different types of AM technologies have been developed for generating STSs, namely, selective laser sintering (SLS), stereolithography (SLA), fused filament fabrication (FFF), direct ink writing (DIW), and inkjet printing. For different printing methods and device requirements, materials need to be carefully considered for the final products.

Creating new materials with new functionalities or creating new functionalities from existing materials are one of the major driving forces for the evolution of 4D printing sectors. Among different types of sensor materials, the magnetoelectric tactile sensor is a newly emerging technology that enables mechanical pressure to be converted into electrical signals with the help of the magnetic field. Wu et al. (6) pioneered the development of a flexible integrated magnetoelectric sensor through the principles of 4D printing. The tactile sensor developed by the group, which exhibits piezoelectric behavior for efficient energy conversion from external pressure, seamlessly aligns with the fundamental tenets of 4D printing. The sensor comprises two parts: a porous structure made via SLS using TPU/NdFeB composite powders, and a helix structure with flat plates fabricated using SLM and 316 stainless steel powders. Magnetic flux changes upon compression of the structure induces electricity via electromagnetic induction. Figure 1 illustrates the fabrication process of the magnetoelectric device.

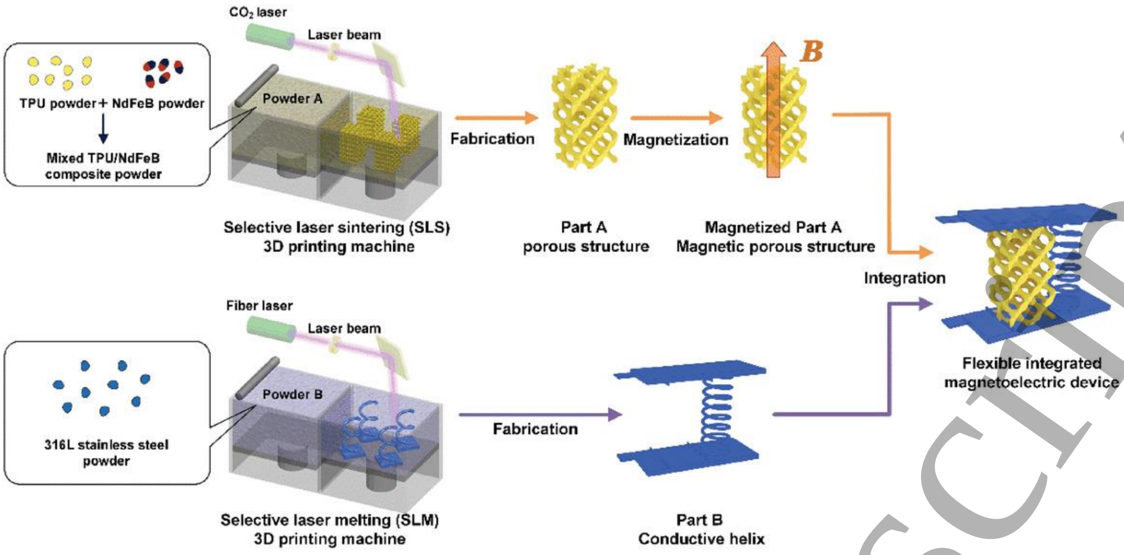

Figure 1. Schematic illustration of the fabrication process of the flexible integrated magnetoelectric device assembled by parts A and B. The part A, a porous structure, was fabricated by selective laser sintering (SLS) additive manufacturing (AM) process using TPU/NdFeB (where, TPU, thermoplastic polyurethane) composite powders and then was magnetized to acquire permanent magnetism. The part B, a helix structure with two flat plates, was fabricated by selective laser melting (SLM) AM process using 316L stainless steel powders. Reproduced from ref.6 with permission of WILEY-VCH Verlag GmbH & Co.

In essence, the convergence of 4D printing and innovative sensor technologies, such as magnetoelectric tactile sensors [6], design-based printed sensors [7], or multifuctional sensors [8] created through 3D printing, holds the potential to revolutionize industries and usher in a new era of responsive and adaptive materials and devices.

Current and Future Challenges

Soft sensors produced through additive manufacturing (AM) have made significant strides, but there are key challenges that must be addressed to fully realize their potential. One of the primary challenges is the limited availability of suitable materials for fabricating soft sensors. Soft sensors require materials that are both flexible and sensitive to environmental changes, which may not align with the capabilities of existing 4D printing materials. Material selection is a critical aspect of soft sensor development. Finding materials that possess the right combination of flexibility and sensitivity remains challenging. The existing materials may not always meet the requirements of soft sensors, necessitating the development of new materials. Another significant breakthrough would be the ability to fabricate an entire soft tactile sensor (STS) in a continuous multi-material 4D printing process instead of printing individual sensor components. This would streamline the manufacturing process and enhance the sensor's overall performance. Integrating multiple materials with different properties, such as materials possessing properties like conductivity and flexibility, is essential for creating effective soft sensors. Achieving seamless integration and proper adjustment of these properties during implementation in 4D printing is complex but crucial for sensor functionality.

Ensuring the durability and long-term reliability of 4D-printed soft sensors is currently another challenging aspect. These sensors must withstand mechanical stress, environmental factors, and repeated use. This will enable consistent and accurate sensing performance in realizing practical soft sensors by optimizing factors like signal-to-noise ratio, sensitivity, and durability for specific applications to achieve the precision and detail required for effective sensing. High-resolution 4D printing with minute features and intricate designs could play a vital role in sensor performance, while this area of research is particularly deficient.

Designing soft sensors for specific applications demands a deep understanding of the intended environment and the collaboration of interdisciplinary fields. Tailoring sensors to various applications, such as medical devices or wearable technology, requires a versatile and adaptable design approach that will meet regulatory standards and ensure safety and reliability as soft sensors find applications in industrial sectors. Last but not least the price needs to be cut to make the actual products available to all.

Advancing 4D printing technology and unlocking its full potential for creating innovative and functional soft sensors will rely on interdisciplinary efforts and ongoing research and development. Overall, addressing these challenges will require collaboration among material scientists, engineers, designers, and medical experts.

Advances in Science and Technology to Meet Challenges

Numerous possibilities could be envisioned to beat the challenges of developing sensors via 4D printing. Advances in materials science are of foremost importance and to addressthese aspects researchers are developing various stimuli-responsive smart materials as well as composite materials for sensor applications. Nanoscale materials and structures are being incorporated into 4D-printed sensors to enhance sensitivity and precision. Nanomaterials like graphene and carbon nanotubes can be integrated into the printing process to create sensors with exceptional performance. To enhance the performance of the sensors, their design-based functionalities are also taken into consideration which has added to the benefit of 4D printing. The work of Mousavi et al. is one such example [7]. They developed a novel tactile sensor using carbon-nanotube-reinforced polylactic acid (PLA-CNT) material. They introduced an innovative approach by creating anisotropic structures for multidirectional tactile sensing. To evaluate the sensor's performance under significant deformation, they adopted a sandwich design, enclosing the PLA-CNT sensor between two layers of thermoplastic polyurethane (TPU) as depicted in Figure 2. The sensor's capabilities were assessed through direct stretching and bending tests, including tensile tests using wooden strips at various angles (0°, 30°, 45°, 60°, and 90°) with a 1% tensile strain applied for three cycles in each direction. The printed sensors exhibited remarkably high sensitivity, with strain gauge factors (k) measuring approximately 1,342.

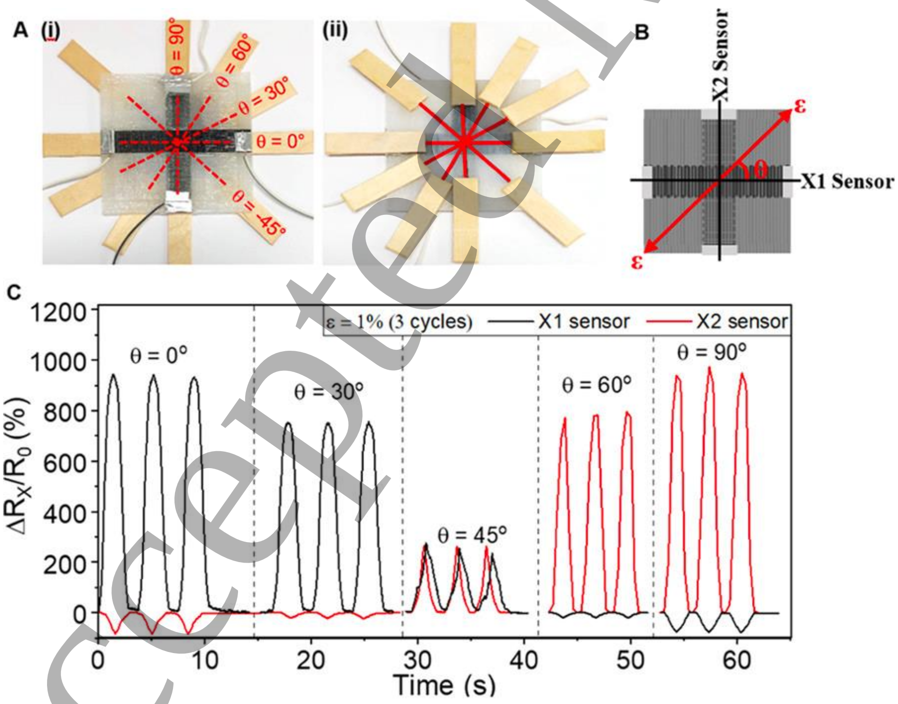

Figure 2. Electromechanical performance of the cross-sensor. (A-i) Front and (A-ii) back pictures of the 3D-printed cross-sensor showing the wooden strips used in tensile tests in different directions. (B) Schematics of a cross-sensor with two X-sensors, namely, X1 and X2, for k_{X1} and k_{X2} calculations. The stretching direction (θ) is also denoted. (C) Tensile strain sensitivity test results in 0, 30, 45, 60, and 90° directions for the 95% infill cross-sensor where a 1% tensile strain was applied for three cycles in each direction. Reproduced with permission from ref. 7.

4D-printed sensors can be designed to serve multiple functions. For example, a sensor could change its shape to optimize its sensing capabilities while also acting as an actuator. Integrating multiple functionalities into a single sensor can reduce complexity and cost. For instance, Chen et al. have created printed integrated sensor-actuators that can respond to thermal stimulation and detect strain through a unique combination of nanocarbon black and polylactic acid [PLA] composites, coupled with bioinspired gradient microgap structures [8]. These sensor-actuators can actively interact with objects upon thermal stimulation and simultaneously monitor their contact state by detecting changes in electrical resistance. This innovative design concept can be applied to develop sensor-actuators with diverse shapes and functions to suit a wide range of applications.

4D printing offers the flexibility to tailor sensors for specific needs, spanning from microscale biomedical sensors to macro-scale physical tactile sensors. Integrating wireless communication will propose better utilization in soft robotics and biomedical fields. In healthcare, there's potential for real-time physiological monitoring, drug delivery, and tissue engineering, with adaptable sensors for biological environments. Furthermore, the fusion of printed sensors with AI and machine learning promises improved data analysis, enabling predictive responses to environmental changes. As 4D printing technology advances, researchers and engineers are actively exploring its potential to revolutionize sensor applications across industries, creating highly adaptive and responsive systems to address diverse challenges and opportunities.

Concluding Remarks

In contrast to conventional manufacturing methods, which suffer from drawbacks such as intricate fabrication processes, inefficient material usage, and limited design flexibility, 3D printing offers distinct advantages for creating smart sensors by efficiently and cost-effectively fabricating sensor components like substrates, sensing elements, electrodes, and dielectric layers in an intricate 3D structure. The sensing functionality of the printed structure thus created the 4D printing-enabled sensors with optimized structural capabilities sensors such as enhanced sensor performance, quicker response times, heightened sensitivity, and improved flexibility with intricate design-based sensing structures. While 4D printing has already demonstrated numerous benefits for sensor production, it is crucial to address certain challenges discussed in this article to fully harness the potential of this technology in the context of 4D printing of soft sensors

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14– 4D Bioprinting

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Status

Since its inception, 3D bioprinting has been a versatile technique for fast fabrication with unprecedented spatial design control; 4D bioprinting is a more advanced version that incorporates higher resolution, dynamic structural, and physiological functionality. 4D bioprinting is essentially a derivative of 4D printing: a term coined by Skylar Tibbit from Massachusetts Institute of Technology in 2013 [1]. The 'fourth dimension' introduced in the 3D printed constructs containing cells encompasses any of the following: a desired transformation of shape, or an alteration in functionality/ property over a desired period, in response to an external stimulus. 4D bioprinting is much sought after in tissue engineering to meet the demands of increasing organ transplantation donors, fabrication of 3D anatomical models, prosthetics, and surgical implants. 4D bioprinting is yet in the proof-of-concept stage, but it has stirred research in the fabrication of complex structures that may serve in tissue engineering of bone, cartilage, nerves, skin, blood vessels, etc,. Polysaccharides have long been recognized as the most adaptable bioinks for 4D bioprinting because they can be easily modified to form responsive bioinks. For example, chitosan has been magneto and thermo-responsive to act as a smart material. [2]. Cell traction force (CTF), or the contractile force exerted by cells, has been explored in order to create cell-origami of 3D structures from 2D laden cell sheets. [3]. Single and bilayered Self-folding tubes have been fabricated, the former based on degree of crosslinking vary in size and diameter. An average internal diameter of 20 μm in a single layered self-folding tube has been fabricated from methacrylated alginate (AA-MM) and hyaluronic acid (HA-MM). A bilayerered structure of alginate beneath poly caprolactone (PCL) was made into a self-folding tube construct, stimulated by a change in the concentration of Ca^{2+} ion. The differential swelling in the anisotropic parts of deformable hydrogels made of alginate (Alg) and methylcellulose (MC) has led to the generation of a variety of complex forms. [4]. The bioink of β-tricalcium phosphate/polylactic acid-trimethylene carbonate (TCP/P(DLLA-TMC)) stimulated by near infrared exhibits deformation, resulting in the formation of a self-folding tube construct. Poly (glyceryl dodecanoic) acrylate (PGDA) has been developed as a novel smart material polymer (SMP) capable of forming complex three-dimensional structures. Silk-GMA (glycidyl methacrylated silk) has been studied in vivo on rabbits and found to have controlled deformability, allowing it to merge well with the native trachea [5]. Servant et al. created a graphene scaffold to facilitate drug release. Carbon nanotubes (CNT) and hydrogen acetate (HA) have also been used as electro-responsive bioinks. [5][4].

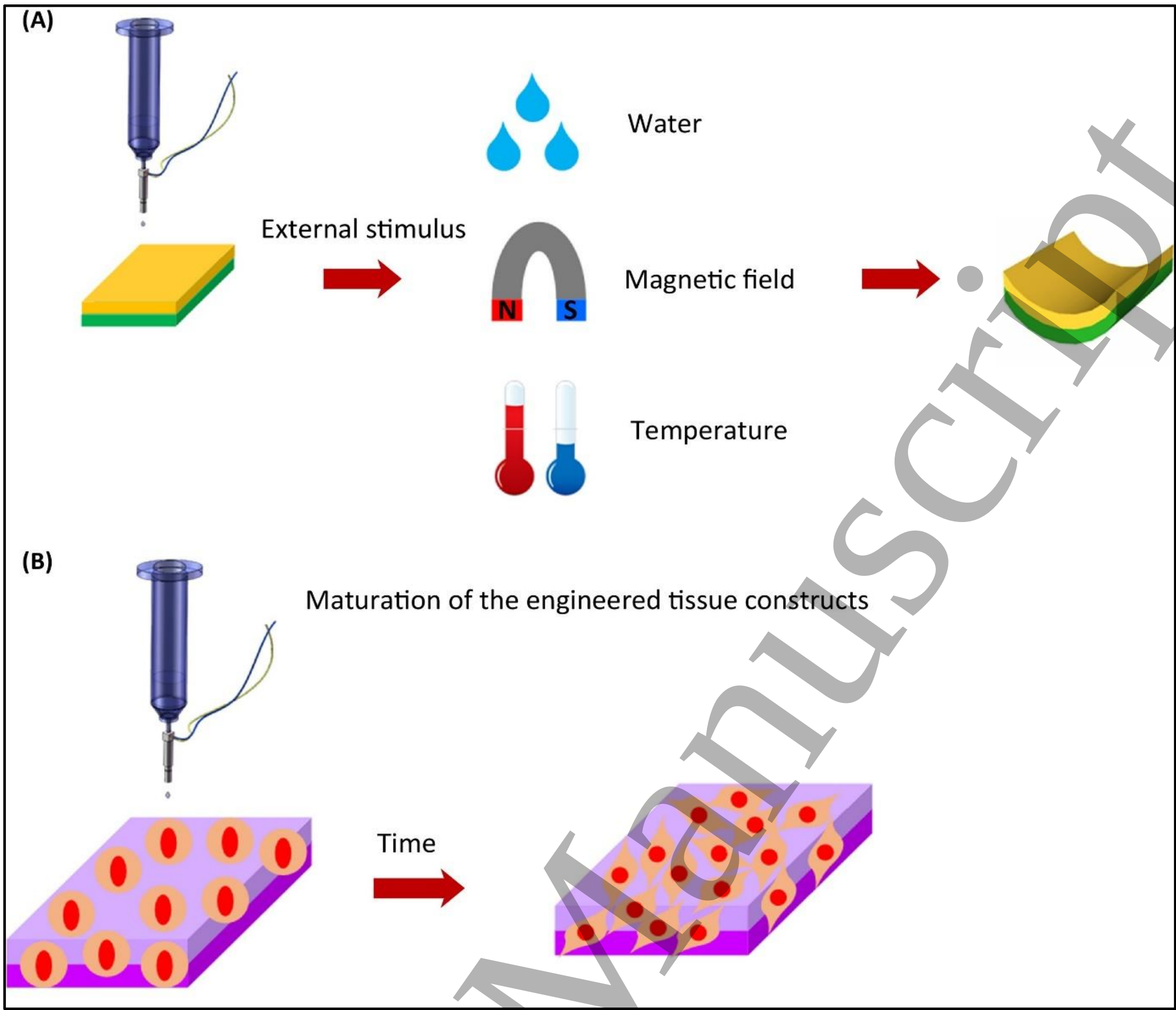

Figure 1. Demonstrates 4D bioprinting [4]via two approaches (A) A material deforming under the effect of stimulus. (B) A 4D printed construct maturing over time. Reproduced with permission from [18].

Current and Future Challenges

4D bioprinting diversifies the use of 3D printed constructs by allowing changes in functionality or structure to occur over time as a response to a stimulus. The challenges faced by researchers in this field include the limited availability of materials and techniques specifically designed for 4D bioprinting [6]. Such materials need to be biocompatible, biodegradable, exhibit good mechanical strength and in addition, must execute dynamic function or structure non-cytotoxically in response to an external stimulus of pH, light, temperature, humidity, acoustics, ions, cell traction force (CTF), electric or magnetic field. Furthermore, 4D bioprinting is bound to shape memory polymers (SMPs), shape memory composites (SMCs), shape memory alloys (SMAs) and composites of hydrogels (SMHs), and conferring biodegradability and biocompatibility on blends of shape memory polymers is not economical yet [7]. Although, SMPs and SMHs are the most common materials used in 4D bioprinting due to their tunable mechanical strength and biocompatibility, SMAs find use in bone tissue engineering. For instance, scaphoid nonunion has been treated with the use of platelet-rich plasma gel and shape memory nails [8]. Those materials that are used for 4D bioprinting are encountered with the challenge of cell seeding which is limited to surface along with non-uniform dispersion inside the matrix of the polymer. Process design is yet another limitation of 4D bioprinting, the design is dependent on the 3D bioprinting techniques that in turn are coping with limitations of resolution, mechanical strength, time-consumption, large-scale manufacturing, and reduced viability due to shear stresses. Although the

4D bioprinting materials are responsive, they still do not closely mimic the living tissues, besides their dynamic properties and the computational models for folding, deformation, bending, and reversal to the initial state after attaining a final temporary shape, are still not well explored. Additionally, the present 4D constructs can only exhibit simpler deformations; including folding, bending, assemblage, curling that do not mimic any of the complex structure of living tissues [8]. Furthermore, most 4D materials are not responsive to more than one stimulus. For humidity-responsive materials, there is no controlled removal of water from the constructs, while the use of salt concentrated solutions for water removal causes osmotic shock to the cells. Besides, swelling, and continuous folding and unfolding affect other properties including mechanical strength leading to poor repeated response to stimuli [9]. Besides, 4D bioprinted model drug constructs still lack specificity, long-durations of drug release and precision.

The future challenges of 4D bioprinting include the introduction of the 4D constructs to clinical applications with extensive trials on materials to improve their properties for stimuli-response, mechanical properties, biocompatibility, biodegradability. A more detailed and comprehensive study of physiological conditions depicting the functional requirements of a tissue or organ needs to progress further.

Advances in Science and Technology to Meet Challenges

4D bioprinting is being explored to form blend bioinks such as a blend bioinks, of gelatin methacryloyl (GelMA) and poly(ethylene glycol) dimethacrylate (PEGDM) has been exploited for structural anisotropy to cause deformation upon hydration [10]. 4D bioprinting has also been researched to cause a 'shrinking effect' post printing to allow generation of thin-scale membranous tissue (TMT) using anionic GelMA and cationic poly-l-lysine (PLL) solutions [11]. The effects of mechanical stimulation in the long-term are expected to be nominal, hence more smart materials that exhibit response to a variety of mechanical stimuli need to be developed. 4D bioprinting has greatly influenced various medical and pharmaceutical industries, it has an enormous potential for bioimplants, bioactuators, stents, biorobotics and tissue engineering [12]. Additionally, the development in fabrication techniques such as multi-material, multi-process printing, hybrid, self-assembly and self-folding and machine learning, and AI driven techniques would advance 4D bioprinting [13]. As the popularization of 4D bioprinting is expanding, it is foreseen to provide new business and commercial opportunities along with a boon to personalized medical industry thus enriching biomedical treatment [7]. 4D bioprinting is garnering attention both in industrial and academics as well, the growth of scientific literature pertaining to the latter has grown exponentially over the past decade (nearly 130%.) along with the rising number of start-ups in this realm. This allows speculation for its overall market to see a potentially tremendous growth soon [14] It is expected by a large group of researchers that by the year 2038 4D bioprinting will lead to drug testing and human disease models, to fit the research based on tissue engineering, while replacing animal testing completely, making clinical trials easier [15]. 3D bioprinting has already been extended to food sector for its scalability and spatio-temporal controllability. However, its popularity may account to an economic loss to the meat sector worldwide and it needs to be verified on ethical and moral grounds of each country. 4D bioprinting in this regard would help to generate complex shapes of various food items, help control their nutritive value and reduce pollution globally [16]. Moreover, commercialization of 4D bioprinting is poorly developed, SWOT (strength, weakness, opportunity, threat) analysis, TRL (technology readiness level) must be evaluated to measure and manage risk due to it, before being utilized commercially [17].

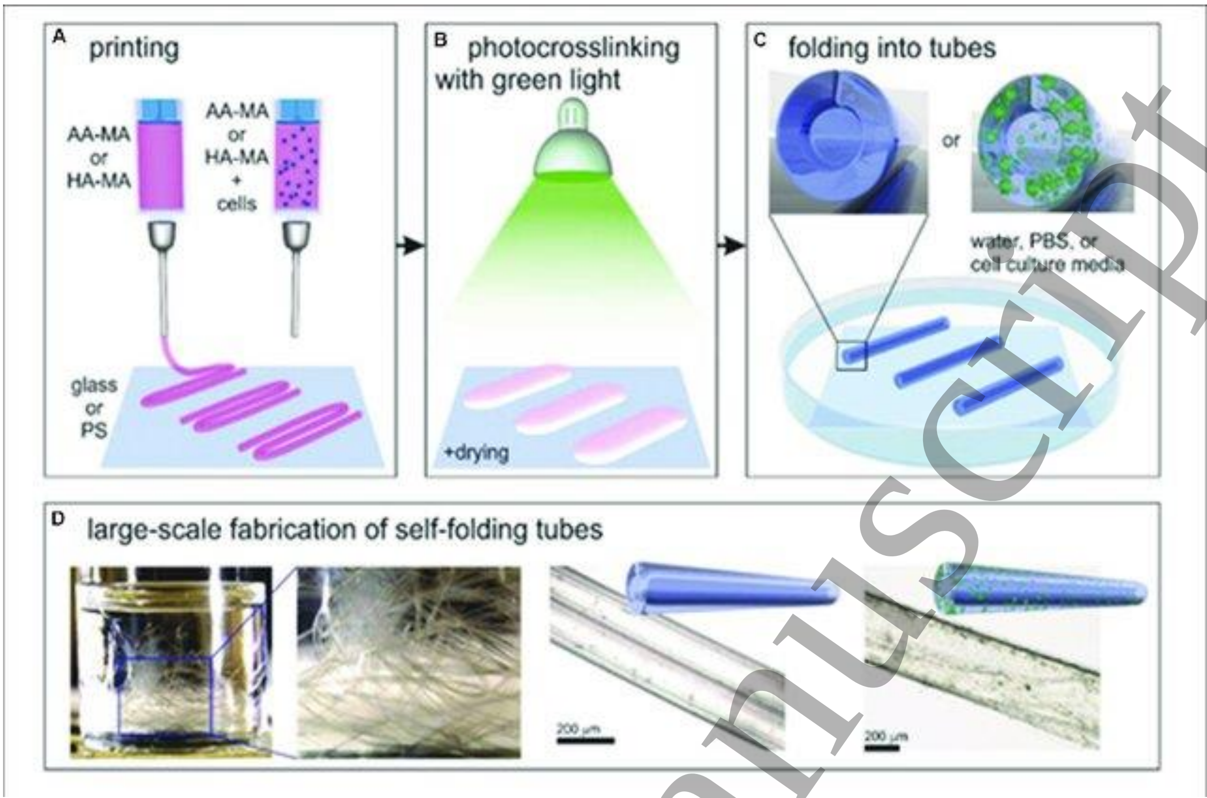

Figure 2. A, B, and C shows the printing of self-folding tubes using two different materials. (D) shows the large scale fabrication of those self-folding materials. [19]

Concluding Remarks

3D bioprinting has demonstrated applicability in engineering tissues, development of prosthetics, surgical implants, and medical devices. However, as the clinical need for transplants is soaring, 4D bioprinting is envisioned as the next generation tissue engineering technique and is believed to replace the functionally and structurally static 3D printed constructs completely. 4D bioprinting also addresses the challenges of fabrication of reduced dimension constructs with higher resolution. This field is also being revolutionized with the development of novel, smart, and responsive materials and the use of machine learning and artificial intelligence, will not just help to treat abnormalities and malignancy in tissues but trauma as well, by in situ bioprinting. 4D bioprinting also fosters the use of small bioactive based material constructs that can incorporate releasing molecules and growth factors as well.

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15- 4D Biofabrication

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Status

4D Biofabrication is an engineering field aiming to develop methods for fabricating structures/objects using living cells and non-vital organic and inorganic materials. This is achieved by harnessing spontaneous transformations, such as maturation and shape changes, in the fabricated living objects (Figure 1) [2]. Shape transformation enables the creation of hollow elements with complex shapes, such as tubes or capsules, or combinations thereof. These are often challenging or impossible to fabricate using standard techniques. Shape transformation is typically accomplished using polymers that can alter their properties, such as hydrogels and shape memory polymers, which allow significant shape changes under conditions compatible with cells [3]. Other shape-changing polymers, such as conjugated polymers and piezoelectric polymers, are used less frequently. Common stimuli for these transformations include temperature (the most widely used) [4], light (often through heat conversion) [5], the concentration of bivalent ions (e.g., Ca2+) [6], or magnetic fields (incorporating magnetic particles) as they are cell-compatible. Other stimuli, such as pH changes, can only be employed in the absence of cells.

4D biofabrication has shown promise in fabricating vascular elements, skeletal microtissues [7], cardiac muscle microtissues [7], and nerve conduits [8]. It encompasses various processing technologies, including photolithography, electrospinning, extrusion-based 3D printing, various forms of light-based 3D printing, direct electrowriting, and others. Typically, shape transformation of individual fabricated objects is employed. However, synchronized shape transformation of two or more objects allows the creation of more complex 3D shapes that cannot be achieved by folding a single object. For instance, two flat objects with specific shapes can fold into a T-junction, which is crucial for fabricating vascular networks (see Figure 2) [9]. Shape transformation enables the transfer of surface patterning from flat objects into 3D patterns. This approach has been utilized, for example, in the fabrication of multilayer structures resembling blood vessels composed of endothelial cells, smooth muscle cells, and fibroblasts [10, 11].

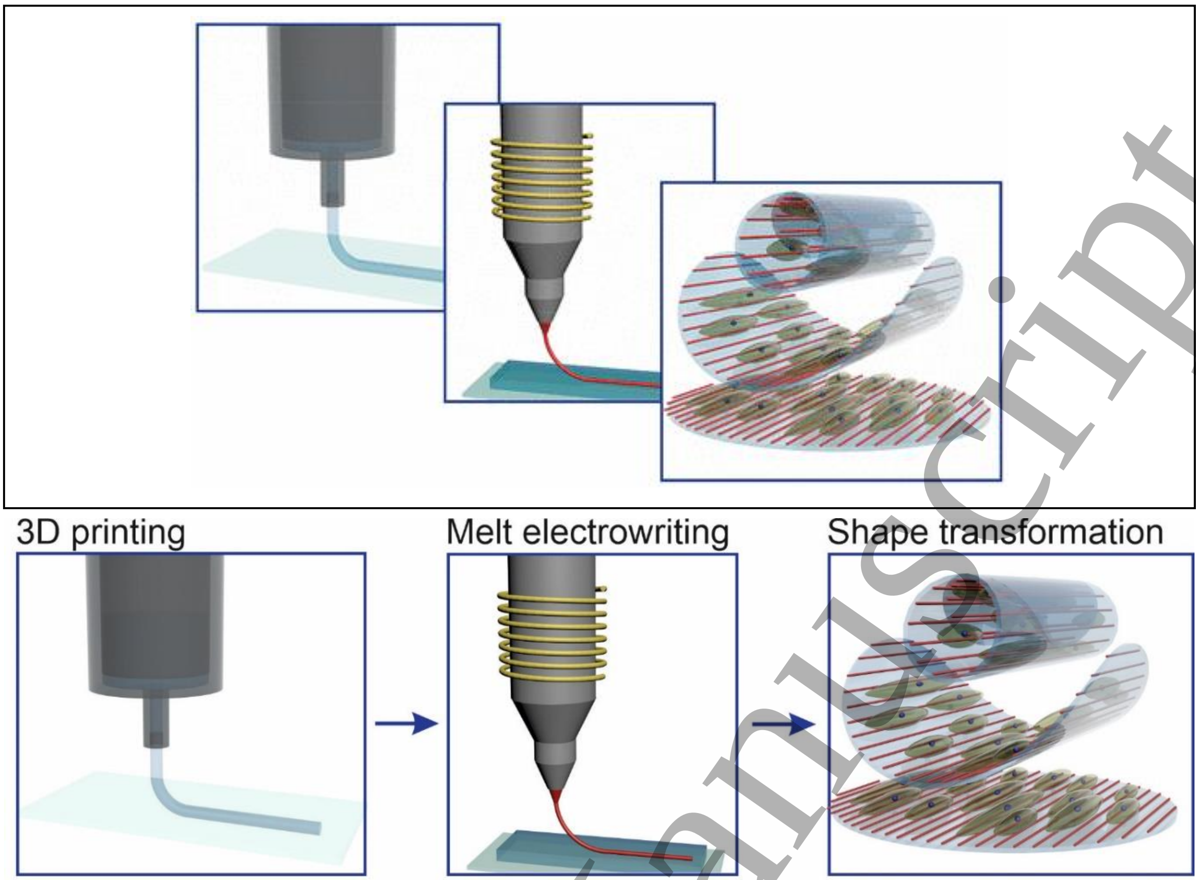

Figure 1. Schematic of the 4D biofabrication process for creating tubular structures with uniaxially patterned inner surfaces. This process involves a combination of extrusion 3D printing with a hydrogel forming solution (left image) and electrowriting of polymer melt (middle image), followed by shape transformation (right image). [1] (Reprinted with permission from ref 1. Copyright 2021, American Chemical Society)

Current and Future Challenges

Despite its promises, 4D biofabrication still faces many challenges that limit its applicability. One of the challenges is the difficulty of combining non-vital materials used for fabrication with living cells to support viability, proliferation, and maturation (differentiation) of cells. Indeed, both natural and synthetic materials are often processed under conditions that are not compatible with cells. Organic solvents, extreme pH levels, high temperatures, or the necessity to dry materials are commonly involved. One bottleneck is how to integrate shape transformation, which is stimulated by external signals, with cell viability. The most 'cell-compatible' stimuli include temperature and the concentration of certain ions such as Ca^{2+}, which imposes considerable restrictions on the choice of polymers suitable for 4D biofabrication. The most versatile stimulus, allowing the use of all polymers, is mechanical force. The easiest way to suppress shape transformation is to apply force, and the easiest way to allow shape transformation is to remove the force. Regardless of the type of stimulus used to trigger shape transformation, one of the technical challenges is the irreproducibility of shape transformation. This issue often arises not only from sample-to-sample variance but also from the viscoelastic behavior of polymers and their long relaxation times. Polymers are not equilibrium materials, and all their properties, including actuation behavior, depend on their prehistory and change over time. Manipulating individual objects obtained through shape transformation, especially if the objects are very small (<1 mm) or soft, is also a significant challenge. The primary potential application of 4D biofabrication is tissue engineering, and one of the challenges is how to provide the proper (bio)chemical and structural environment for cells to direct their development in the right ways.

Advances in Science and Technology to Meet Challenges

Future research directions in the field of 4D biofabrication must encompass the resolution of current challenges, exploration of new possibilities, and the development of innovative approaches. Moreover, it necessitates a profound investigation into the interactions between materials and cells, cell development, and tissue formation both in vitro and in vivo. For instance, addressing the challenge of insufficient reproducibility, a critical factor in advancing targeted 4D biofabrication, entails the comprehensive collection and analysis of all data related to samples, their structure, and their history.

One potential new direction in 4D biofabrication involves the use of multiple shape-changing objects. While the shape transformation of individual objects is akin to the Japanese art of paper folding, origami, the synchronized transformation of several objects resembles modular origami. For example, the synchronized folding of two layers with specific shapes allows for the creation of T-junctions, crucial elements in vascular networks that cannot be fabricated through the folding of a single layer.

A significant bottleneck in the commercial application of biofabrication is the need for extensive biological studies, including animal experiments, due to their extended duration and high costs. Once these challenges are overcome, we can anticipate the utilization of the 4D biofabrication approach for producing microtissues as alternatives to animal experiments. Scaling up, which becomes technically feasible once the problem of reproducibility is resolved, will further reduce the costs associated with fabricating tissues. This, in turn, may lead to the application of 4D biofabrication in the field of cultured meat engineering.

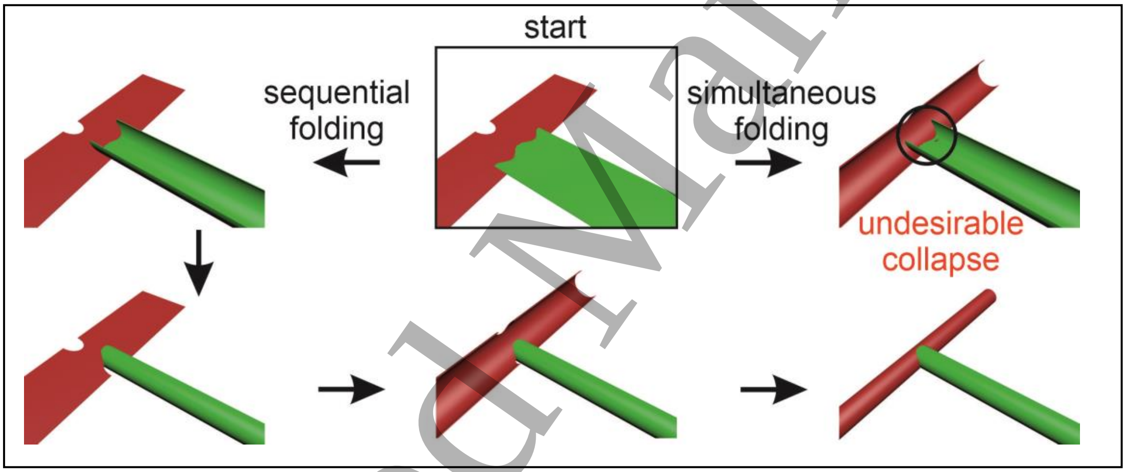

Figure 2. Example of fabrication of complex 3D shapes by synchronized folding of multiple shape-changing objects.

Concluding Remarks

Despite significant strides in recent times, 4D biofabrication still grapples with numerous unsolved challenges, notably the issues surrounding the reproducibility of shape transformation and the limited depth of biological studies. These challenges constitute the foremost priorities that must be tackled in the immediate future to pave the way for the industrial application of 4D biofabrication, particularly in crucial domains like tissue engineering and biotechnology, including the burgeoning field of cultured meat production.

One of the most pressing short-term objectives within the field of 4D biofabrication is the development of innovative approaches. For instance, the exploration of novel techniques for fabricating intricate structures using multiple objects is an avenue ripe for investigation. This approach resembles modular origami, where synchronized transformations of several objects result in the creation of complex structures that are otherwise challenging to achieve with a single object. Such innovations not only expand the capabilities of 4D biofabrication but also hold immense potential for breakthroughs in tissue engineering and biotechnology applications.

The future of 4D biofabrication hinges on overcoming these hurdles and leveraging innovative strategies, propelling the field towards a future where it can revolutionize the way we produce tissues, organs, and even sustainable food sources like cultured meat. As research and development efforts intensify, we can anticipate remarkable advancements and a broader impact on various sectors of science and industry.

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16– 4D printing in cancer therapies

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Status

Cancer has been known for a long time; however, the understanding of cancer and its treatment remains complicated. It is the cause of millions of deaths globally, which is one in six deaths, and there is still a tendency to increase [1-4]. As seen in Figure 1(a), in history, cancer has been almost cured by surgery to remove the cancer cells, followed by radiotherapy and chemotherapy to obtain effective outcomes and prevent recurrence. New cancer therapies have been continually explored. 4D printing is one of the new technologies mentioned in cancer-therapeutic studies. It has been emerging in the fabrication of smart structures used in cancer therapeutics, such as scaffolds [5-7], devices [5, 8], and robots [9-12] (as shown in Figure 1(b)), to address the issues of personalization [13], controllable dose [14, 15], and site-specific area of treatment [11]. One further significant reason that 4D printing is suitable for cancer management is the unique environment of cancer, which results from its abnormal metabolism. To be more explained, the pH value of the cancer site is more acidic than that of the normal site, and some enzymes like matrix metalloproteinases (MMPs), and hyaluronidase (HAase) show upregulated levels around the cancer sites [4, 9]. These factors could be used as biomarkers and stimuli for inducing 4D-printed structures. Since 4D printing techniques have various manufacturing methods and smart materials, the understanding and insight of printed structures have been realized. There are some summaries about 4D printing for breast cancer management and an overview of 4D printing in cancer therapeutics studies [3, 4]. However, the research on 4D printing in cancer treatment is still limited. Therefore, a lot of information is still needed to progress and succeed in animal trials or clinical trials. The research project of 4D printing in cancer therapeutics still involves anticancer drugs, including programmable locomotion, drug models, and controllable drug release. For example, the fabrication and use of multipurpose implants loaded with drugs for breast cancer treatment were recently revealed [13]. It is also possible to push these revealed protocols into clinical trials by enhancing the biocompatibility of materials.

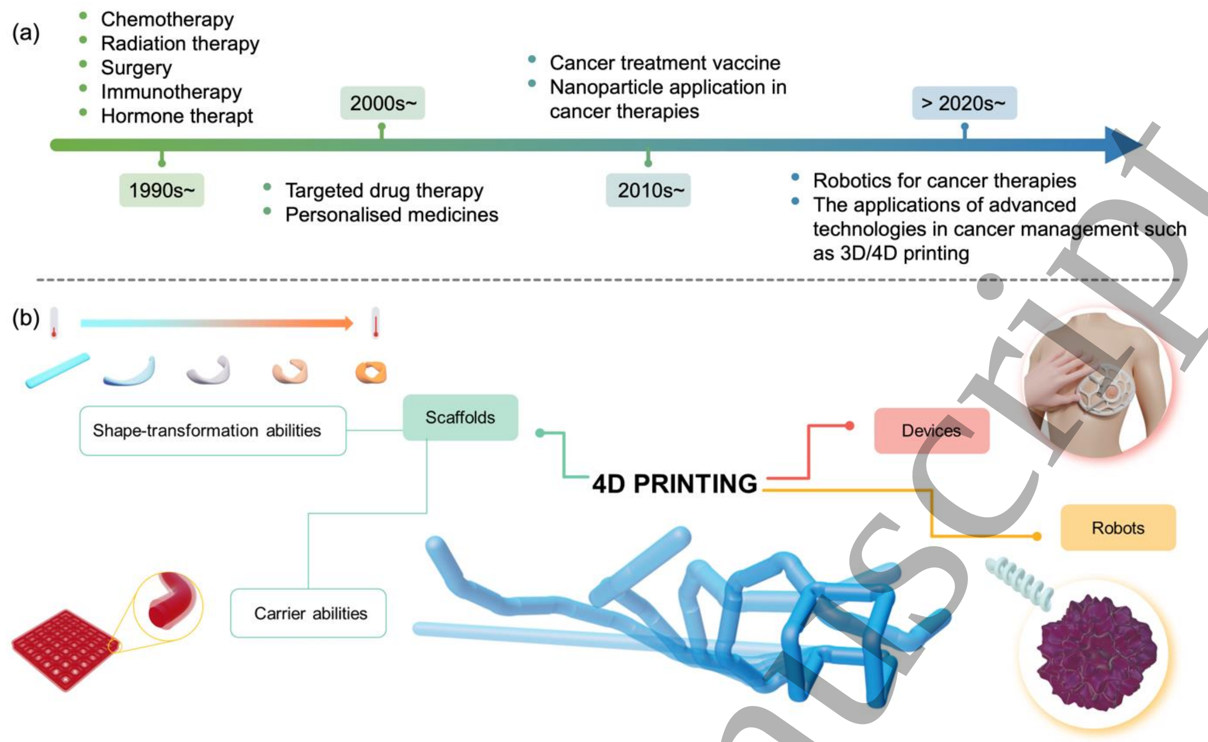

Figure 1 (a) The brief history and future trends of cancer treatment, and (b) an overview of currently dynamic products in cancer therapies that were fabricated by 4D printing.

Current and Future Challenges

Several studies have reported the use of 4D printing techniques to fabricate smart structures in cancer therapies. 4D printing has been used to provide personalized scaffolds, patterned structures, and multifunctional abilities. For example, Chen, et al. [16] revealed the abilities of a smart tracheal stent that was able to perform self-expanding for the patient's trachea fitting, control the Paclitaxel (PTX) drug for chemotherapy, and generate heat under an alternating magnetic field (AMF) for hyperthermia, however, challenges remain. First, stimuli employed in 4D printing procedures are needed under physiological conditions. For instance, the pH used to drive dynamic microrobots with controllable anticancer drugs is approximately 7.4 [12]. Second, there are many procedures that are needed to be investigated, such as imaging, biodegradation, undesired immunoreactions, doses, side effects, or post-treatment symptoms, before applying them to an *in vivo* experiment or clinical trial [6, 9, 17]. As seen in Figure 2, all steps of the clinical trial of smart biodegradable microswimmers were envisioned. The microswimmers based on magnetic hydrogel, which were fabricated by two-photon polymerization, could perform controllable locomotion, biodegradation, and drug delivery. Hence, navigation in the body under an external magnetic field, released dose, degradation under the pathological microenvironment, and medical imaging should be assessed.

In addition, another important challenge of 4D printing in cancer treatment is the improvement of protocols to be applicable with new therapies. 4D printing has been applied to fabricate smart structures used in some therapies such as surgery, chemotherapy, hyperthermia, targeted therapy, and combination therapy, even though its advantages have not been mentioned in many therapies such as immunotherapy, especially advanced therapies like stem cell therapy, nanoparticles, natural antioxidants, and gene therapy. To maximize the benefits of new cancer therapies and evaluate the potential of 4D printing, new strategies for its application in these cancer treatments will be revealed in the future.

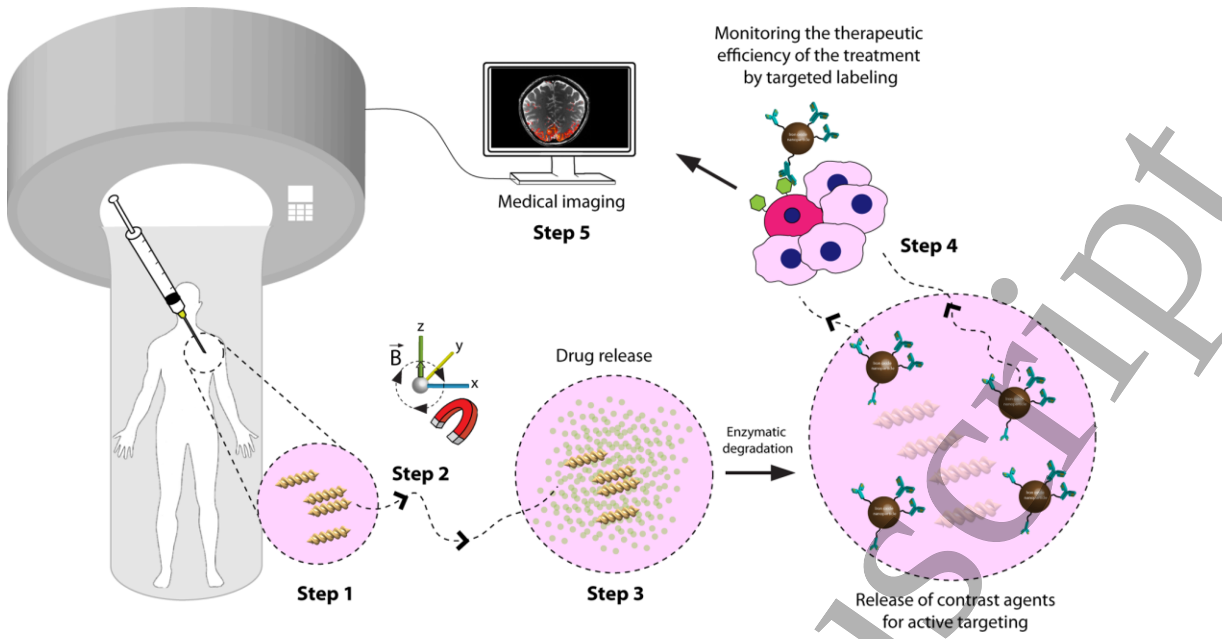

Figure 2 The envisioned situation of smart microswimmers, printed by a direct laser writing, for cancer treatment. Reprinted from Ceylan, et al. [9]. CC-BY license.

Advances in Science and Technology to Meet Challenges

As mentioned in the previous section, the cancer therapies have been improved because there are still some concerns with conventional therapies, such as severe toxicities to normal cells, local residual cancer, recurrence, and tissue loss [15]. Nanoscale structures, including nanoparticles and nanomedicine, have been presented to reduce the risk of nonspecific distribution and side effects [18]. The fabrication procedure of dynamic nanostructures by using 4D printing has required improving the printing resolution or combining with other methods to achieve the nanoscale. For instance, a tetrapod microstructure, which was one of the parts of a sperm-hybrid micromotor, was fabricated by two-photon 3D nanolithography to guide and release the sperm loaded with an anticancer drug locally for the target drug delivery system [11]. The nanostructures not only enhance the potential of cancer treatment but also expand the range of possibilities for accessing hard-to-reach areas [9, 10]. So, nanofabrication technologies such as a two-photon polymerization-based femtosecond laser 3D printing technology [18] or the combination of digital light projection 3D printing and nanoscale-relief patterning [19] are possible to apply in the 4D printing procedure to fabricate dynamic nanostructures for cancer therapies in the future.

Besides, 4D printing relies on many sections, including printing procedures, smart materials, and structural changes, which led to extensive time spent studying and analyzing multiple data sets. There were studies of machine learning used in predicting tumors and cancer treatments. The machine learning modelling might be useful to predict the changes in smart structures on 4D printing in cancer management in future research [20].

Concluding Remarks

4D printing has emerged as an important research tool for cancer therapies. The fabrication of dynamic structures that provide customized structures and programmable characters has attracted significant interest. 4D printing can provide personalized medicine and treatment, which is highly desired in cancer treatment. 4D printing has been utilized in the fabrication of multifunctional scaffolds, programmable devices, and controllable robotics used in cancer therapies such as chemotherapy, surgery, thermotherapy and hyperthermia. However, applications for 4D printing in new cancer treatments, such as immunotherapy, and stem cell therapy, and clinical trials are still lacking. In addition, the fabrication of 4D-printed structures to realize the potential of cancer therapies has been continually developed. The nanoscale fabrication and machine learning of 4D printing have been mentioned as ways to advance cancer therapies through 4D printing research.

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17 - 4D Printing for Pathophysiology

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Status

Pathophysiology is a domain that describes and studies abnormal changes in a biological function, encompassing the causes and/or consequences of an illness. Studies of pathophysiology cover several disorders, such as cancer, cardiovascular diseases, wounds, and neuro-injuries, by investigating their origins and treatment [1]. Over the years, various approaches have been explored to improve local therapy. 4D printing has emerged as a path to creating custom-made complex structures that mimic biological tissues, such as implants or drug delivery systems, that respond to a certain stimulus. 4D-printed devices can interact with the adjacent environment, change their conformation, and trigger different biological responses. Consequently, shape-changing materials (SCM) combined with additive manufacturing technologies have been pointed out as solutions for pathophysiology management [2]. Table 1 displays some of the SCMs that are suggested in the literature.

Table 1. Some Examples of shape-changing materials that can be applied to pathophysiological applications of 4D printing (Adapted from [3]– [5]).

Type of Stimuli	**Materials**	**Application**	**Disorders**
Temperature	Poly(lactic acid) (PLA)	Self-expandable stents	Coronary artery disease
	Polycaprolactone (PCL)	Self-expandable stents	Tracheal stenosis
			Coronary artery disease
	Polyurethane (PU)	Paracetamol-releasing implants used in knee replacement	Bone or joint replacement
	Poly(N-isopropyl acrylamide) (PNIPAM)	Drug release system	Several disorders
	Poly (D,L-lactide-co-trimethylene carbonate) (PDLA-TCM)	Personalized intravascular shape-changing stent	Atherosclerosis
	Poly(vinyl alcohol) (PVA)	Gastro-retentive and intravesical drug delivery systems	Gastric disorders
Magnetic	PLA/Fe_3O_4 composite	Shape-changing stent	Tracheal stenosis
Electrical	Fibrin Protein blended with poly(acrylic acid) (PAA)	Drug delivery systems based on the mobility of ions	Several disorders

Light	Poly(ethene glycol diacrylate) (PEGDA)	Cardiac patches	Cardiovascular diseases
	Hydrogel of alginate + gelatine (core) and Polydopamine (PDA) + PCL (shell)	Drug release system	Cancer
pH	Alginate	Wound dressing that detects bacterial biomarkers and heals skin infection using a drug delivery system.	Skin wounds
	Hypromellose acetate succinate	Drug delivery system	Several disorders
	PAA	Drug delivery system	Several disorders
Biological Stimuli	Hyaluronic acid	Cardiovascular scaffold with improved neovascularization	Cardiovascular diseases
	Gelatine	Drug delivery system	Several disorders
	Crosslinked Alginate and F127DA hydrogels	Drug delivery systems	Several disorders

Currently, self-expandable stents, drug delivery systems, and wound dressings are the most commonly described 4D-printed systems in the field of pathophysiology. Due to the wide range of possible applications, the most explored are 4D-printed drug delivery systems, which still present scientific gaps to fill. Considering the materials, there is a demand to find SMCs that respond to internal changes in pH, enzyme activity, and glucose uptake because these stimuli indicate metabolic abnormalities. Many researchers foresee that in the following years, 4D-printed smart hydrogels combining cells, polymers, and other responsive materials will play a key role in pathophysiology, namely in patient-centric therapeutics. However, converting novel 4D-printed structures into an efficient product will be challenging and time-consuming.

Current and Future Challenges

4D printing has already proven valuable in areas like biorobotics, biosensors, tissue engineering, and organ transplantation [6][7]. However, as it is a very recent strategy, it offers a wide range of research opportunities to pursue technological advancements. Only a very few studies have focused on the application of 4D printing in pathophysiology [8]– [10]. One of the main reasons is the lack of understanding about which pathological conditions can serve as biological stimuli and how they will influence the smart materials' properties. For instance, pathologies are commonly associated with unbalanced biochemical metabolisms translated into ion concentration changes; accordingly, studies on both physiological changes in ionic strengths and ion-responsive materials design could improve awareness of the benefits of this stimulus in pathophysiology treatment.

The most representative challenge relates to the reversibility of printed structures. 4D printing describes a reversible conformational change over time [6][11]. Therefore, 4D printing must involve SCMs that resume their original shape when the stimulus is removed. After the stimulus is removed, the challenge relies on adequately implementing SCMs and also exploring the influence of the recoverability property of SCMs on biological structures. In this context, some authors refer to the development of multi-stimuli-responsive materials as a promising prospect in pathologies because of the multiple microenvironmental changes in diseased tissues (temperature, pH, redox potential) that could possibly be used as biological stimuli [8].

The challenge of reversibility can be surpassed by 4D bioprinting. By reshaping cell morphology over time, 4D bioprinting can controllably induce bio-tissue development [12]. Even though 4D bioprinting still requires intense investigation, it can be vastly adopted in the future. Low scalability, long fabrication times, and heat and/or radiation resulting from 3D printing technologies still hamper the widespread application of 4D bioprinting.

Simultaneously, since not all stimulus-responsive materials can be used in 3D printing, another challenge relates to the development and upgrade of 3D printing technologies, enabling a wider range of materials to be applied in 4D printing for pathophysiology. Similarly, exploring new smart materials based on available 3D-printable ones could expand the possibilities of 4D-printed system design.

Finally, an imminent challenge is the expansion of the 4D printing paradigm to the nanoscale. As the pathophysiology occurs at the biochemical level, the nanoscale is more suitable for its management. Nevertheless, before this enlargement, more studies addressing the improvement of the 4D printing precision should be conducted to allow the use of nanomaterials.

Advances in Science and Technology to Meet Challenges

The use of 4D printing in the healthcare sector has received much attention in the last few years, with a compound annual growth rate (CAGR) of 26.7% and an estimated market size of USD 32 million by the end of 2026, rising to USD 36 million in 2028 [13]. Because healthcare is such a broad field, in this section, the focus is on the advances needed to meet challenges in the field of pathophysiology through biomedical implants, tissue engineering, or therapeutic delivery (Figure 1).

3D printers can build devices with special and complex features in a short time, permitting the manufacture of low-stability pharmaceuticals for fast intake and custom-made devices wherever the spatial-temporal conditions demand urgent action, like in intensive healthcare units, surgery rooms, military facilities, or ambulances. Thus, the pathophysiology field will benefit from specialized and accurate printers to place custom-made products safely in the body. For that, 4D-printed drug delivery systems need to pass rigorous criteria for clinical applications, for instance, high selectivity and accurate and prompt responses to stimuli inside the body. Also, it is necessary to upgrade and control the release profile of drugs in the targeted environment according to the selected stimulus [14].

Regarding the printing of cells to help tissue regeneration or patient-specialized therapy (e.g., cancer), it is necessary to evaluate the cell viability decrement influenced by the printing technique. Changes caused by the laser, heat, radiation, or enforced shear or mechanical stresses may alter the biological performance of printed cells. Printing parameters like nozzle diameter, layer thickness, or porosity must also be considered since they can induce cell damage or decrease the mechanical properties of the medical system [14].

In respect of biomedical implants to treat diseases (e.g., atherosclerosis, malformations, respiratory tract diseases), they may favor an intense study of the influence of printing parameters on the mechanical properties, biomimicking performances, and therapeutical efficiency and, thus, be adequate for each patient [15]. One possible way to overcome some issues is to use artificial intelligence (AI) to simulate and predict the modeling and stimulation of medical systems [3].

Lastly, there is a need to correspond to the FDA requirements for medical products and demonstrate the strong efficacy and safety of the 4D-printed designs, particularly for commercialization, real-time applications, or scaling-up production.

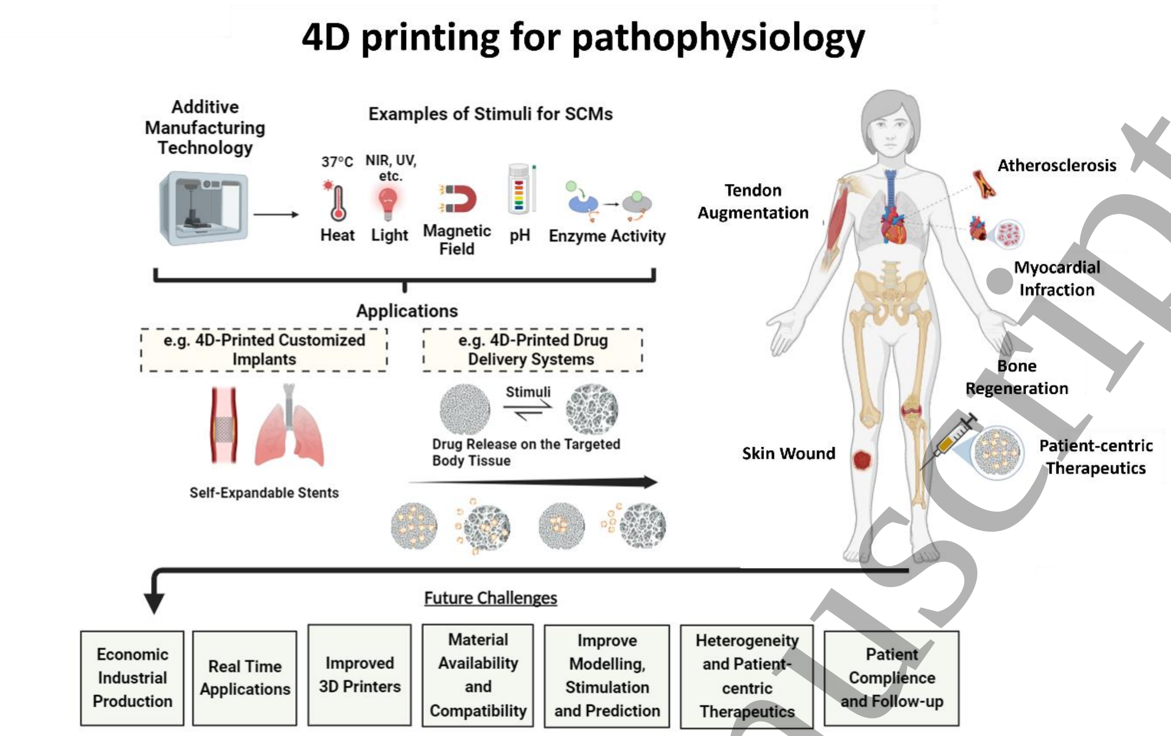

Figure 1. Schematic representation of future challenges of 4D printing for pathophysiology (Created with BioRender.com).

Concluding Remarks

The advent of 4D printing has enabled the broadening of horizons concerning pathophysiology management. This is mainly due to the combination of two critical factors: the capability of printing custom-made devices or implants, and the ability to change the conformation upon stimulation with an adequate environmental stimulus. Indeed, as the pathological surroundings are characterized by physiological and homeostatic changes of temperature, pH, and ionic concentration, among others, they constitute themselves good candidates to be used as internal stimuli and, therefore, induce conformational modifications over time. The major challenge lies, however, in the correct and efficient use of such triggers to promptly handle the pathologies. Therefore, open and interdisciplinary communication between physicians, biochemistries, and engineers with different competencies and backgrounds will be essential for designing, upgrading, improving, and implementing innovative 4D-printed systems in pathophysiology.

Acknowledgements

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18 – Micro- and Nano- 4D Printing

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Status

While 4D printing at the macroscale is experiencing a prosperous development, the demand for precise micro- and nanostructures with adaptable properties has exponentially increased in innovative areas such as biomedicine, micro-optics, and micro-robotics. Thus, new approaches enabling the precise manufacturing of smart objects at smaller scales became essential. Among all the available 3D printing techniques, light-based 3D printing has the great advantage of producing objects with intricate geometries at high resolution and high speeds.[1] In particular, two-photon laser printing (2PLP) allows for manufacturing on a nano-meter scale with high precision.[2] In brief, this technology utilizes a femtosecond-pulsed high-energy laser for locally exposing a photo-reactive material in 3D and inducing spatially confined two-photon polymerization. Due to the non-linearity of the process, resolutions well below the micron are achieved. Currently, this technology is being commercialized by several companies. However, the development of new functional materials to achieve the "fourth dimension" using 2PLP is still in its early stages.[3] Major efforts have been recently made in this regard in the design of printable stimuli-responsive materials. In the following sections, current state-of-the-art materials and technologies for 4D printing at the micro- and nano-scale along with their challenges and future perspectives, will be summarized.

Current and Future Challenges

Material design

For the fabrication of smart 4D micro- and nano-structures, 2PLP has been established as the preferred technique. Therefore, the development of suitable formulations for two-photon polymerization is one of the key steps. Typically, they are composed of three main components: stimuli-responsive monomers, crosslinkers, and a two-photon initiator. During the last few years, promising examples of defined 4D microstructures employing hydrogels, liquid crystals, shape memory polymers, and composite materials have been shown.[3] For example, stimuli-responsive printable hydrogels based on acrylamides, specially *N*-isopropylacrylamide, and/or acrylates have been successfully employed for the preparation of thermo-responsive soft microactuators (Figure 1A).[4, 5]. Liquid crystalline-based materials have also been exploited as excellent candidates for the preparation of micro-actuators (Figure 1B) and tunable optics.[6–8] In addition, microstructures exhibiting a shape memory effect (Figure 1C),[9, 10] and magnetically powered micro-swimmers for application in drug delivery have been reported (Figure 1D).[11]

While the potential of 4D microprinting has already been successfully demonstrated, there is a pressing need for continued endeavors to integrate this concept into practical real-world applications. Consequently, forthcoming research on effectively controlling the response of the printed structures at smaller scales is essential. For instance, novel materials with the capability to autonomously respond to multiple stimuli with spatially controlled responses will greatly enhance the versatility of these systems in applications in the area of micro- and nanorobotics. Furthermore, biocompatibility, biodegradability, and integration with biological systems are highly desired features in the biomedical area.

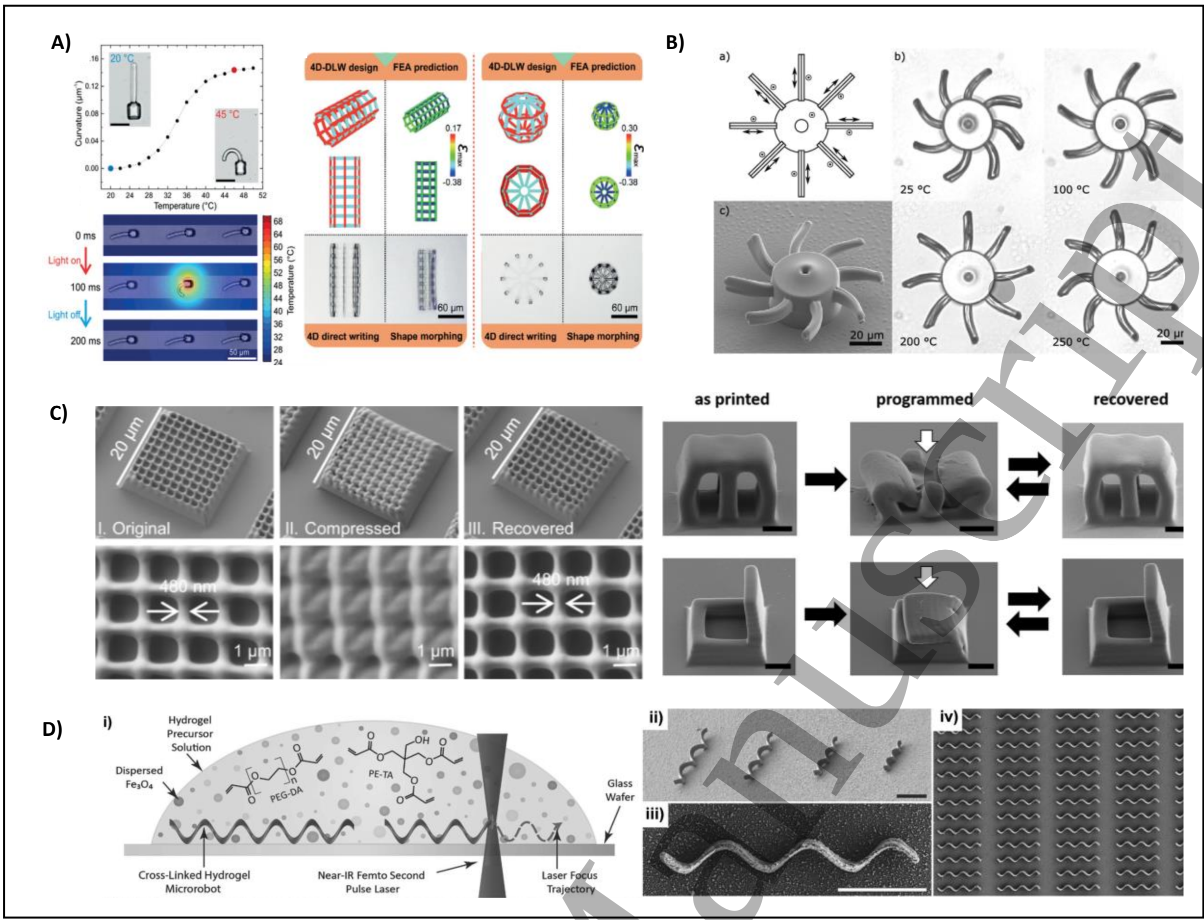

Figure 1. Recent examples of 4D microstructures: A) Thermo-responsive poly-NIPAM microactuators (Left panel adapted from [4] Copyright © 2019, The Author(s); right panel adapted from [5] © 2019 The Authors. Published by Elsevier Ltd.), B) Liquid crystalline 4D microstructure ((Adapted from [6] © 2021 The Authors. Advanced Materials Technologies published by Wiley-VCH GmbH), C) shape memory 4D microstructures (Left panel adapted from [9] Copyright © 2021, The Author(s); right panel adapted from [10] © 2022 The Authors. Advanced Functional Materials published by Wiley-VCH GmbH) and D) magnetic microswimmers (adapted with permission from © 2015 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim).

Technology

While two-photon laser printing (2PLP) possesses the significant benefit of enabling high-resolution 3D printing, this technology also has some drawbacks. One of them is the high cost of the system due to the necessity of an expensive femtosecond laser. Additionally, the printing process can be rather long, especially when large areas or amounts of printed structures are targeted. In other words, scalability is still one of the challenges that must be tackled in 3D/4D micro-printing. Recently, major efforts have been made in parallelization [12] as well as alternative processes, which will be discussed in the next section.

Advances in Science and Technology to Meet Challenges

In order to continue towards the success of 4D printing at small scales, further advances in both material science and technology are essential. To this aim, future efforts relying on multi-disciplinary approaches must be taken. As mentioned above, one important consideration for future applications, especially biomedical applications, is biocompatibility. Further work in the search for non-toxic monomers and photo-initiators is essential. In addition, integrating full biodegradability would allow, for example, easy implementation of printed systems in biomedicine. Biodegradability would also minimize the impact of these future materials on the environment, decreasing plastic waste and pollution. Furthermore, moving towards "life-like" 4D materials that enable not "just" response to external stimuli but great adaptability and customized design in regard to geometrical and mechanical, physical and/or chemical

properties will be a key point in future applications. For example, our group recently presented a promising approach combining 2PLP with dynamic covalent chemistry and "living" polymerization that enables the fine-tuning of the size and mechanical properties by several degrees of magnitude on-demand.[13] Thus, we believe that by integrating all the above-mentioned features – biocompatibility, biodegradability, and "life-like" behavior – in printable materials, new dimensions can be reached.
At the technological level, new 3D printing approaches that rely on different photo-initiation mechanisms have recently emerged as alternatives to traditional two-photon processes. These mechanisms include triplet-triplet annihilation, triplet fusion up-conversion and two-step absorption processes [14–17]. Importantly, these methodologies do not require large and expensive femtosecond lasers but enable precise 3D printing using just compact and low-cost illumination sources. Although these techniques are still in their infancy, a future perspective would be their implementation in 4D printing. By designing 4D printable materials with the features discussed above, new opportunities will emerge.

Concluding Remarks

Over the past few years, exciting advancements in materials and technologies for 3D and 4D printing at small scales have been made. Despite the challenges outlined, the burgeoning field of 4D printing at the micro- and nanoscales holds great promise. Smart and dynamic microstructures, whose properties can be precisely programmed and controlled as needed, will exert a significant impact in the very near future in several disciplines ranging from micro-robotics to optics and biomedicine.

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19 - High Resolution 4D Printing

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Status

4D printing is a cutting-edge manufacturing technology that integrates 3D printing with smart materials to create 3D structures that can change their shapes or functions over the fourth dimension – "time" in response to external stimuli such as heat, moisture, electric/magnetic field, or others. Such shape-morphing structures can be printed by various 3D printing technologies including fused deposition modeling (FDM), digital light processing (DLP), direct ink writing (DIW), Inkjet printing, selective laser sintering (SLS), stereolithography (SLA), and two-photon polymerization (TPP) (Figure 1a). Among them, the high resolution 3D printing techniques such as TPP and DLP enable high resolution 4D printing that fabricate shape-morphing 3D structures with feature size at the micro- and nanoscales [1]. As shown in Figure 1b, TPP technology can fabricate intricate 3D structures with an exceptional printing resolution ranging from 90 nm to 500 nm, relying on the two-photon absorption (TPA) theory. This process involves the simultaneous absorption of two photons from near-infrared (NIR) light at equal low frequency (v/2), lifting a molecule from its ground state to a higher energy level to induce polymerization in photosensitive resin [2]. Different from TPP that fabricates 3D structures by scanning the nanoscale laser spot through two photo absorption, as shown in Figure 1c, DLP technology creates 3D structures by projecting ultraviolet patterns through a digital micromirror device (DMD) to on the surface of photosensitive resin. The feature size of the projected pattern can be improved by optimizing the optical lens, and the highest resolution for DLP can be as small as 0.6 μm [3]. Both TPP and DLP can be used to achieve high resolution 4D printing by printing photopolymerizable smart materials including hydrogels [4], liquid crystal elastomers (LCE) [5], shape memory polymers (SMP) [6-8], magnetic/electric responsive materials [9].

Owning to the capability of seamlessly integrating multifunctionalities into complex 3D structures with micro- or nanoscale features, high resolution 4D printing can generate intelligent microstructures and microdevices, which have found extensive applications in diverse fields including mechanical metamaterials, flexible electronics, micro-robotics and biomedicines. Shape-transformable structures [6] (Figure 1d) and even mechanical metamaterials [7] (Figure 1e) can be achieved through DLP printing of SMPs. By printing an ionic conductive polymer with a dielectric, flexible sensors with multimode sensing capability can be achieved [9] (Figure 1f). Multimaterial DLP 3D printing enables multimodal mobile microrobot with piezoelectric building blocks [10] (Figure 1g). The microrobots can be further scaled down to micrometers, which are fabricated by TPP and used for biomedical applications [11] (Figure 1h). Using TPP to print SMP structural colors, tunable photonic devices can be achieved [8] (Figure 1i).

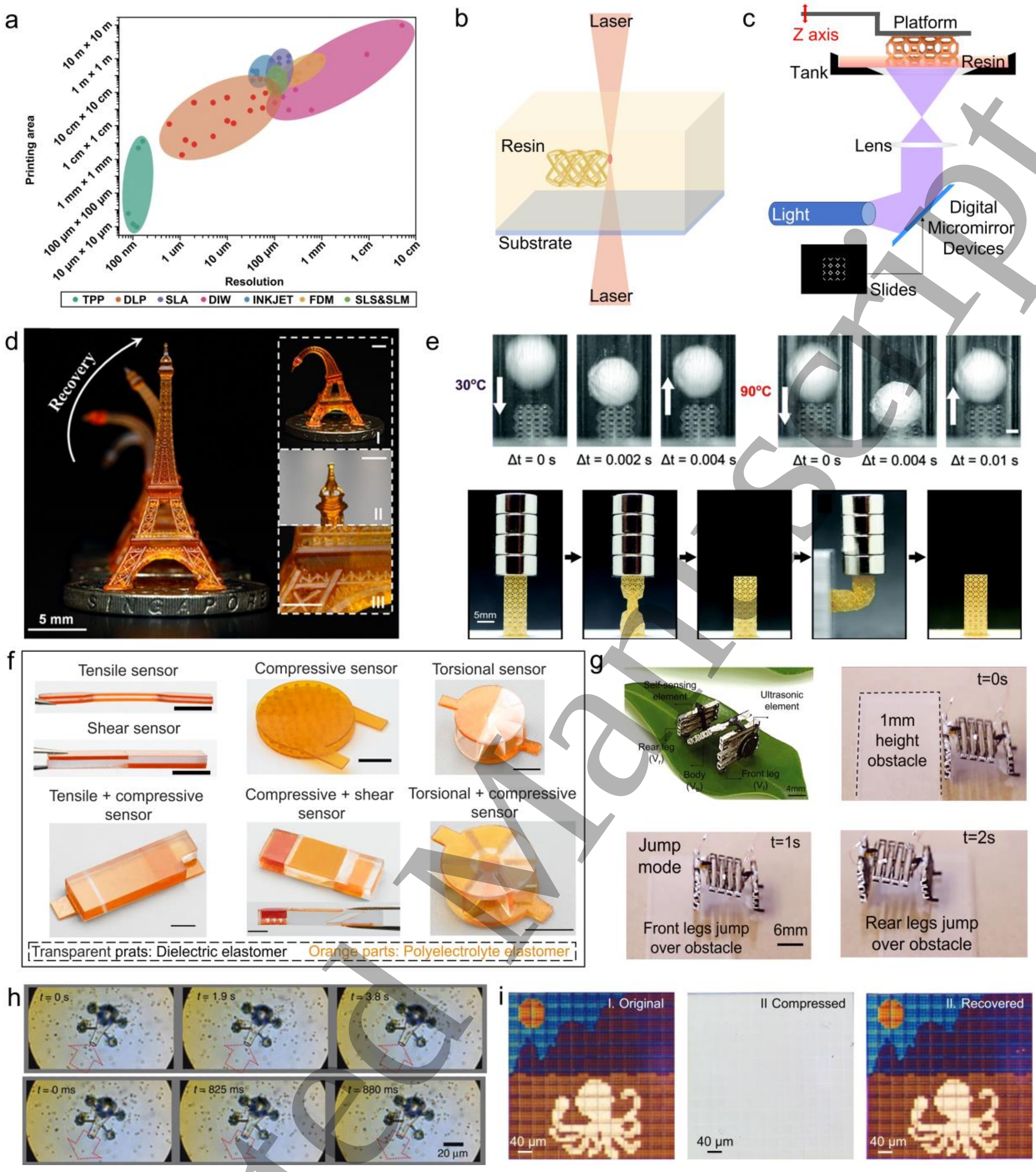

Figure 1. The technical characteristics and typical applications of high resolution 4D printing. a. The diagram summarizes the printing resolution and printing area relation of TPP, DLP, SLA, DIW, inkjet, FDM as well as SLS&SLM [1]. b-c. Schematic diagram of two-photon polymerization and digital light processing. d. High resolution 4D printed shape memory polymer Eiffel Tower [6]. e. Thermally tunable and reconfigurable microlattices [7]. f. DLP 3D printed various architected ionotronic sensors for multi-mode sensing using polyelectrolyte elastomers with robust interfaces and without leakage [9]. g. DLP printed piezoelectric multimodal mobile microrobot climbing obstacles driven by electric fields [10]. h. TPP printed micro-tools eject the trapped particles as the light-controlled piston through the microbubble [11]. i. Shape memory effect of 4D printing structural multi-colour painting [8].

Current and Future Challenges

Despite the recent rapid advances, the implementation of high resolution 4D printing into practical applications is still hindered by the following three key challenges: the conflict between printing resolution and efficiency, limited capability of printing multimaterial structures with refined features, and inadequate smart/functional materials for high resolution 3D printing.

TPP enables the fabrication of shape-morphing structures with feature sizes of 90 to 500 nanometers [2]. Most of these structures were fabricated on a commercial TPP 3D printer (Nanoscribe, Germany), which has the finest planar resolution of 160 nm and vertical resolution of 0.1-5 μm. Although the maximum building volume is 100 × 100 × 8 mm^3, the scanning method to print 3D structures requires about 1852 days to form a bulk volume in such size at the maximum speed of 625 mm/s. Different from TPP, DLP generates 3D structures through mask projection which can quickly form a planar pattern through one shot of projection. The commercial DLP 3D printer (BMF, China) uses projection micro stereolithography to fabricate 3D structures, and its finest planar resolution is 2 μm and vertical resolution is 5 μm, but the projection area is limited to 3.84 mm × 2.16 mm. The step-repeat method extends the projection area to 50 mm × 50 mm, but the full cover of such area requires 312 repeat projections.

Additionally, the current intelligent microstructures and microdevices manufactured by high resolution 4D printing are mainly in the form of single material. This is because that both the printing process in either TPP or DLP occurs in liquid photosensitive resin, and multimaterial 3D printing requires switching of resins, which has not yet been achieved by commercial TPP and DLP printers. Despite many attempts to endow TPP [12] and DLP [13, 14] with multimaterial 3D printing capability, the current systems suffer from small building size, limited applicable materials, and low degree of structural or functional integration.

Moreover, since both TPP and DLP rely on photopolymerization to realize 3D printing, the liquid resins that are converted to the solid counterparts are required to have high photosensitivity and low viscosity. These strict requirements severely constrain the number of smart materials such as SMPs, hydrogels, ionic conductive polymers, and polymer embedded with magnetic/piezoelectric particles that can be printed by TPP or DLP.

Advances in Science and Technology to Meet Challenges

To meet the above-mentioned challenges, significant progress for high resolution 4D printing has been achieved, driven by three key advancements: the high-efficiency, high resolution and large-aera 3D printing, the ability of multimaterial structure printing, and the development of advanced smart/functional materials for high resolution 3D printing.

The combination of multiple printing methods offers a solution to balance both printing resolution and efficiency, thereby enabling the seamless integration of high resolution, large-scale, and high-speed printing within a single printing process. Researchers have proposed several feasible approaches to achieve this goal. Typical examples include the large area projection microstereolithography approach, which combines DLP with a coordinated optical scanning system [15], the femtosecond projection two-photon lithography technique that integrates TPP with the DMD [16], as well as integral lithography, which combines DLP with a rotational microlens array [17].

Efforts have also been made toward achieving multimaterial printing in high resolution 4D printing. Researchers have developed various multimaterial 3D printing devices to tackle the key technical obstacle of efficiently and quickly switching and removing the residual resin. Multimaterial switching can be achieved by switching multiple resin barrels [14], or dynamically controlling fluids through resin switches [13]. Particularly, an innovative DLP-based centrifugal multimaterial 3D printing via spinning technique has been reported [18], enabling the creation of large-volume heterogeneous 3D objects. Additionally, the integration of DIW with DLP technique has greatly expanded the range of available materials and improved the efficiency, versatility and complexity of printed structures [19].

The notable advancements in developing and integrating advanced materials significantly enhance the responsiveness and functionality of 4D-printed microstructures. Introducing novel molecular designs, such as double networks and intermolecular forces, into smart materials can yield robust mechanical properties [20]. The incorporation of nanoparticles into smart materials allows for finer control over their responsiveness [20]. Furthermore, the integration of process-material-structure-function design is essential for the advancement of high resolution 4D printing technology. Utilizing advanced multi-physics modeling and simulation tools has led to a deeper understanding and accurate prediction of the behavior of smart materials during 4D printing.

Concluding Remarks

In the above sections, the status, current and future challenges, as well as recent advancements in high resolution 4D printing technologies, were comprehensively reviewed. The development of high-efficiency, large-area, high resolution, multimaterial printing systems, and advanced smart materials is poised to be applied in various industries. First, as 3D printing devices advance with higher resolutions, larger print areas, and faster printing speeds, we can anticipate groundbreaking developments enabling the creation of highly detailed, large-scale 4D-printed objects with exceptional efficiency. Industries like aerospace and architecture will benefit from the capability to rapidly produce intricate, full-scale prototypes and components. Second, the achievement of multimaterial high resolution 4D printing will usher in an era of versatile and intricate 4D-printed structures. This capability will enable researchers to print materials seamlessly with diverse properties, resulting in functional, multi-modal, and highly adaptable objects. These innovations will advance biomedical devices such as customizable implants, drug delivery systems, tissue engineering, and flexible electronics with intricate, multi-material circuits. Furthermore, the potential applications will be limitless as smart materials advance, exhibiting finely tuned properties that respond to various stimuli. We can envision 4D-printed structures that autonomously self-assemble or morph in real-time in response to environmental changes. These innovations will contribute to soft robotics with intricate design, high responsiveness, and high adaptability.

Acknowledgements

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20 - Accelerated Design For 4D Printing

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Status

The rapid advancement of 4D printing has primarily been fueled by researchers involved in the fields of materials science, mechanical modeling, and chemistry. Most studies focus on tuning, depositing, and stimulating active materials used in various additive manufacturing (AM) techniques. They involve mastering the creation of multifunctional objects and structures while comprehending the interactions between active materials and energy stimuli that give rise to sensing, transduction, and actuation functions [1,2]. Expanding active material performance and deposition capabilities represents a key research strategy, essential before industry adoption. However, harnessing the potential of smart materials and AM to engineered objects capable of adapting, transforming, or deploying in response to environmental or artificial stimuli poses a significant challenge from both design and engineering perspectives [3].

Recent research efforts have been concentrated on multi-material 4D printing, a fabrication strategy that combines passive and active materials to enhance actuation performance and achieve complex shape changes [4,5]. In this context, the successful integration of dissimilar materials within a single object necessitates advancements in the capabilities for depositing multiple materials and computational design techniques. From a design standpoint, precise material distribution becomes paramount for achieving the desired shape changes. An approach using voxel-based modeling has emerged to facilitate property and material assignment within the 3D design space, enabling forward predictions of shape change of printed structures when exposed to any energy stimuli [6]. Furthermore, significant strides have been taken in the inverse design of shape-changing structures. To determine material distribution effectively, researchers have employed a combination of genetic algorithms and machine learning (ML) techniques [7-9], even coupled with topology optimization (TO) [10], resulting in efficient computations on reduced 3D model composed of up three distinct materials (see Figure 1a). These results provide sophisticated arrangement of materials in the 3D design space and are relevant for complex shape-changing behaviors and shapes where engineering experience is not sufficient. However, these voxel-based multi-material structures may encounter limitations with current AM techniques. This printability issue has been addressed by combining multiple AM techniques within a single machine [4,5,11,12], and more recently by introducing interlocking blocks that can be easily printed separately and then assembled. For the latter, a computational design approach has been developed to convert voxel-based material distribution into multiple interlocking blocks made of active and passive materials, leading to promising mechanical and actuation performance (see Figure 1b,c) [13,14].

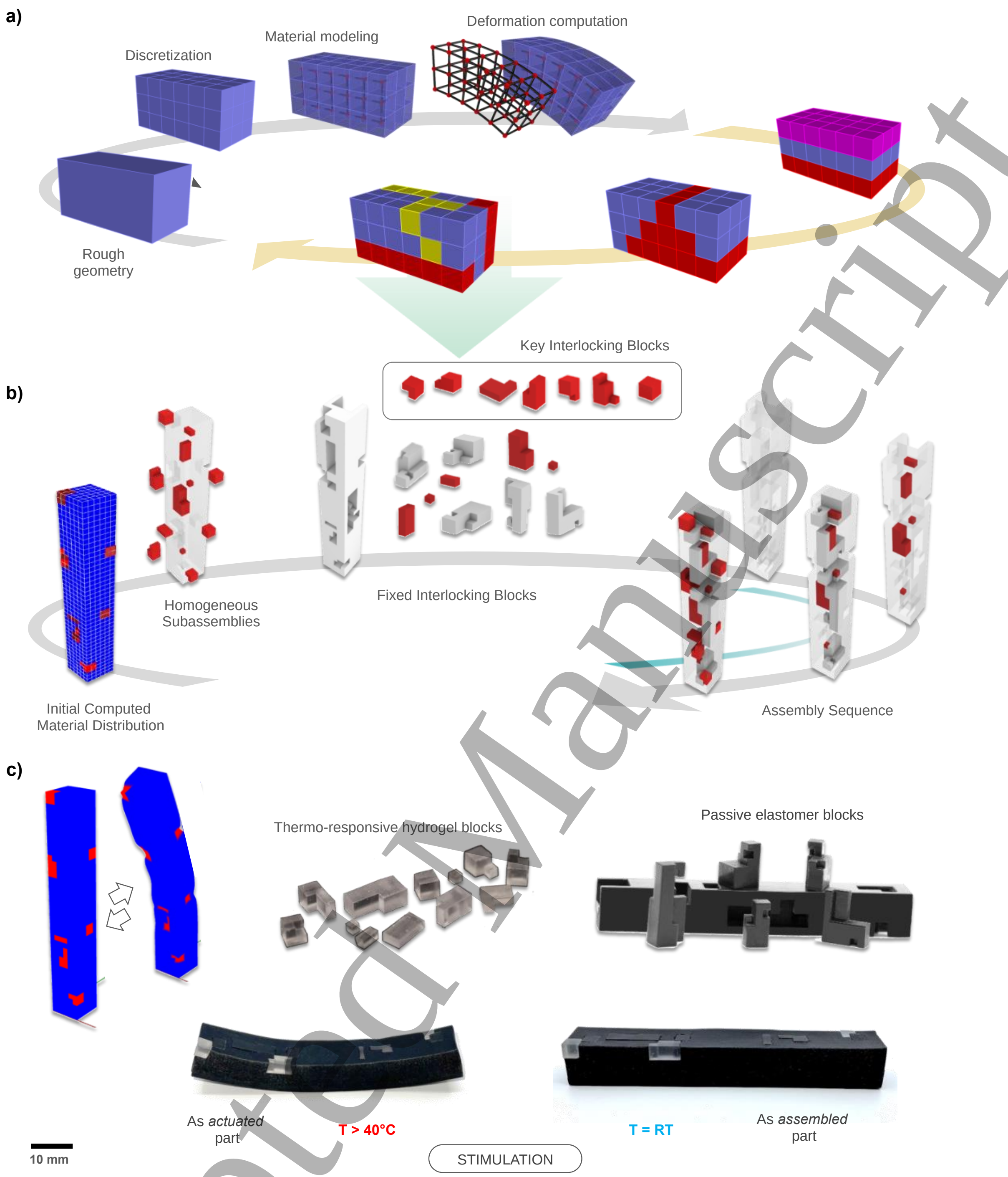

Figure 1. Inverse design strategy and computational approach to build multi-material objects and structures using voxels and interlocking blocks. (a) voxel-based modeling for predicting smart material behaviors; determination of material distribution using genetic algorithms and ML techniques, (b) computational design of complex material distribution into interlocking blocks, and (c) printed active and passive blocks and assembly strategy towards thermally sensitive shape-changing objects.

Current and Future Challenges

In addition to the ongoing research focusing on material distribution in the 3D design space, a systemic perspective is crucial for 4D printing design. The long-term objective is to create adaptive, deployable, and transformable systems, applicable across sectors like medicine, automotive, textiles, aerospace, space, and more. These systems must make practical sense for widespread adoption. A direct challenge lies in capturing hidden needs, stemming from the often-misunderstood nature of 4D-printed objects. Addressing this can expand potential disruptive ideas.

Designing for 4D printing involves concurrent consideration of various factors, including functions, smart materials, expertise domains, worlds, AM techniques, scales, temporal aspects, physics, and stimuli (Figure 2). This comprehensive integration challenge demands increased research efforts to develop models, methods, and software that incorporate computational and reasoning capabilities for various abstraction levels. These tools are vital for rapidly exploring potential solutions (shape, structure, materials, and stimuli) and identifying optimal designs for long-term sustainability. Such challenges fall into two subfields: design *for* X (X representing AM, material assembly, transformation, and performance) and design *with* X (X representing smart materials, stimuli, and artificial intelligence - AI). In the first category, X implies an objective to ease, while in the second one, X means an opportunity for inclusion and enhancement.

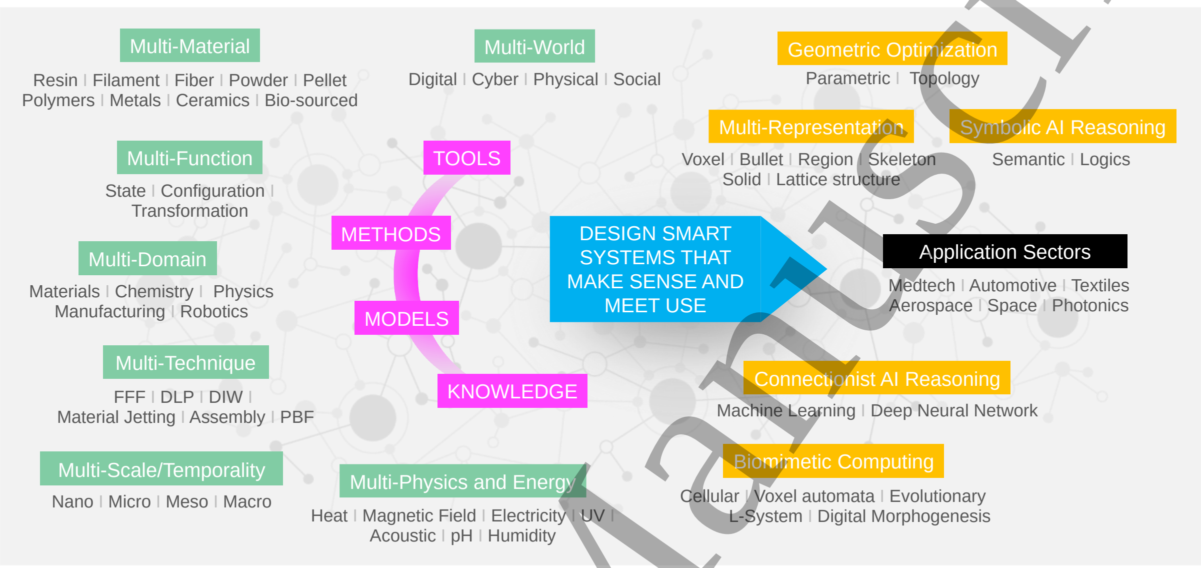

Figure 2. Design for 4D printing roadmap built upon knowledge, models, methods, and tools (in purple boxes) that integrate 4D printing factors (in green boxes), and dedicated reasoning layers (in orange boxes) to meet application sectors (adapted from [3]).

The traditional design process is composed of several phases, starting with design thinking and the creative generation of ideas and concepts, followed by the embodiment and detail design stages. Therefore, it is important to support creative strategies while introducing constraints (objectives and opportunities) to converge towards a satisficing solution. Creativity is essential for 4D printing to establish functions and solve problems at hand and could benefit greatly from biomimicry and sustainable development. Conversely, the embodiment design phase, receiving significant attention in 4D printing, requires more scientific contributions. This includes exploring material distribution at different scales, complex structures, and spatiotemporal stimuli distribution. To achieve this, the use of biomimetic computing with ML and deep learning techniques, supported by extensive datasets, is relevant and cost-effective. However, this strategy requires explanatory capabilities. Leveraging multi-domain ontologies, knowledge graphs, and integrating physical laws, even using physics-informed neural networks for small datasets, is imperative [15,16]. This integration aligns with neuro-symbolic AI, applicable to forward prediction and inverse design, through voxel-based simulation, finite elements simulation, or discrete TO. To accelerate 4D printing design, another challenge is streamlining knowledge, constraints, and decisions across design stages, ensuring digital design and fabrication pipeline.

Advances in Science and Technology to Meet Challenges

The scientific challenges associated with accelerated design for 4D printing involve expanding the research domains related to this emerging technology. This expansion includes considerations of AI, industrial design, and even the utilization of nano-microscale manufacturing technologies. These research domains can be used to manage complexity and introduce novel architectural freedom. Considering the Hype Cycle that highlighted 4D printing as an innovation trigger in 2018 [17], this research field has now reached a typical phase of disillusionment. It is crucial to materialize concrete applications to reach a productivity plateau leading to market adoption. To achieve this, it is important to leverage symbolic AI techniques to expand knowledge bases related to active materials, systems comprising structure-stimuli-materials, and possibly biomimicry but not limited to. This knowledge can be effectively integrated into design and simulation models and methods to tune, justify, validate connectionist AI reasoning, and even serve as a basis for recommendation systems (materials selection, stimuli selection, mechanism selection, etc.) [18,19].

The integration of multiple perspectives in design is challenging and cannot be addressed by a single research group. It is essential to establish scientific networks, whether through funded research programs, or relevant scientific communities in conjunction with industrial partners. From an organizational point of view and to work in a highly pragmatic manner, in the short- and mid-terms, this involves conducting bottom-up actions (from researchers and research groups towards scientific communities and research agencies) to fuel and invigorate the creation of knowledge, materials, and technologies. It also involves top-down operations (from societies towards researchers) to structure collective intelligence and actions towards technological solutions that are beneficial for society and industry [1,2].

Concluding Remarks

Design for 4D printing plays a crucial role for advancing the widespread adoption (society, industry) of this emerging technology. It served as the convergence point for various scientific perspectives, integrating design *for* X (including AM, material assembly, transformation, and performance) and design *with* X (including smart materials, energy stimuli, nature-inspired principles, and AI) strategies. This engineering phase requires a reimagining from a paradigmatic and computational standpoint to accelerate the exploration and discovery of innovative real-world applications and their practical usage.

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21 - Pattern-Controlled 4D Printing

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Status

Shape programming is a critical part of four-dimensional (4D) printing that can be achieved using multiple approaches, such as printing-process-induced programming and post-printing programming. Shape programming can be used to control the permanent or temporary shape of the 4D-printed structure, depending on the type of the programming process [1]. One of the critical factors that influence the shape-programming process is the printing patterns, leading to a diverse area of pattern-controlled 4D printing. This is because the induced strain levels in 4D-printed structures are affected by the paths of these patterns. Pattern-controlled 4D printing can be realized using nine categories of patterns that are summarized in Figure 1, where each one of them offers different levels of complexity and control to the actuation process.

The most common pattern-controlled approach is achieved by manipulating the infill patterns, which are usually generated using standard three-dimensional (3D) printing slicing software [2]. The shape and density of the infill patterns influence the actuation performance of 4D-printed structures. Gradient patterns offer the ability to gradually change the thickness, size, shape, and properties of the pattern [3]. The next type is hierarchical patterns, which are made up of smaller patterns that are repeated in a specific way to create complex structures [4]. Lattice patterns consist of repeating geometric shapes that are used to create objects that can deform or change shape predictably [5]. Auxetic patterns are special types of lattice patterns that possess a negative Poisson's ratio. This means that such patterns expand transversely when longitudinally stretched while offering mechanical advantages, such as energy absorption [6]. Origami and Kirigami patterns are some of the most interesting and complex 4D printing patterns. Origami patterns rely on folding the structure, while Kirigami patterns involve folding structures that include slots as well [7]. Weaving patterns are commonly used in textile-based and hygroscopic-based 4D printing to control the stiffness of the structure at selected locations [8]. Bioinspired patterns mimic patterns that exist in nature to achieve complex actuation performance [9]. Lastly, voxel-based patterns rely on discretizing the structure into sub-blocks using multi-material printing [10,11].

Current and Future Challenges

The main challenge that currently exists in 4D printing is the incomplete understanding of the fundamentals of this technology since it is a relatively new research area. More specifically, pattern-driven 4D printing drastically increases the levels of complexity in terms of design, modeling, simulation, printing, and characterization. The challenges begin even when using the simplest pattern since it is required to perform geometrical modeling of the induced-strain levels within a single shape or component of the pattern. This step becomes more challenging when dealing with non-uniform patterns. Most reported models that aim to estimate the curvature or bending angle of 4D-printed structures rely on variations of Timoshenko's model, where the transient response can be estimated for simple structures [12]. Applying this concept to pattern-controlled structures is a complex task, especially when combined with time-varying stimulus levels or stimulus-rate-dependent responses.

This issue might be addressed using finite element analysis (FEA) approaches that incorporate constitutive models [13].
To date, most researchers in this area focus on using standard computer-aided design (CAD) tools, which can be ineffective when using complex non-uniform patterns. Then, the complexity of the simulation process is affected by the type of patterns used as well as the nature of the printing process itself. Therefore, even FEA faces difficulties when simulating pattern-controlled 4D-printed structures due to the complexity of solving the induced-strain levels along the patterns' paths. Another challenge in this area is the limitations of the slicing software used to prepare the design for the printing process. Despite providing multiple infill pattern geometries, the level of control of the parameters of the patterns is still limited, especially when considering adaptive and gradient infill patterns. Moreover, the printing process suffers from limitations in terms of the dimensional accuracy, repeatability, reproducibility, and scalability of the printed structures, especially when using consumer-grade printers. Even when using industrial-grade printers, the adoption of 4D printing technology by industry is still limited since this technology is not mature enough to be implemented on a large scale without extensive research and development processes. Advancements in pattern-controlled 4D printing require collaboration between academia and industry to tackle the challenges in this area from different aspects.

Advances in Science and Technology to Meet Challenges
It is envisioned that the future development of this area will involve four main steps, as illustrated in Figure 2. The first step starts by defining the design requirements in terms of the actuation performance requirements, material properties, and the stimulus type and range used to activate the structure. The energy required to activate the structure and the self-sensing ability are factors that need to be taken into consideration. Then, the printing requirements are considered in terms of minimizing the use of material and reducing the cost while achieving the desired performance and printing accuracy. The next step, which is design development, starts by selecting suitable materials from an open-access smart materials (SMs) database. It is envisioned that the development of four-axis to seven-axis printers will grow rapidly in the future [14]. These printers will open doors for four-axis to seven-axis 4D printing, which allows complex structures and shape-programming approaches to be realized. This concept will offer the ability to print high-degree-of-freedom patterns that cannot be printed using three-axis printers. As a result, slicing tools need to adapt to these changes using parametric pattern-based slicing approaches. Artificial intelligence (AI) will play a crucial role in the development of this area in terms of AI-enhanced and generative AI design. Moreover, CAD scripting and visual programming language (VPL) design approaches [10,15] can address the current limitations of standard CAD tools. Furthermore, the improvements in the simulation capabilities will see great improvements in terms of developing optimal FEA tools that can solve complex pattern-controlled design problems that might even rely on supercomputers.
The third step involves implementing the design based on the selected printing parameters, patterns, and shape-programming method [16]. This is followed by the assembly and integration of the structures. Lastly, the fourth step implements corrective measures to optimize the whole process. This may involve using computer vision (CV) to detect printing inaccuracy, which can be then reported to a server using cloud-connected (CC) printers. Moreover, 3D scanners can be used during and after the printing process to verify the accuracy of the printed structures. Then, the information can be stored in a 4D printing database that can be used to optimize future designs. The actuation performance is then analyzed and compared with the design requirements, while important information is sent to the second and third steps for further improvements.

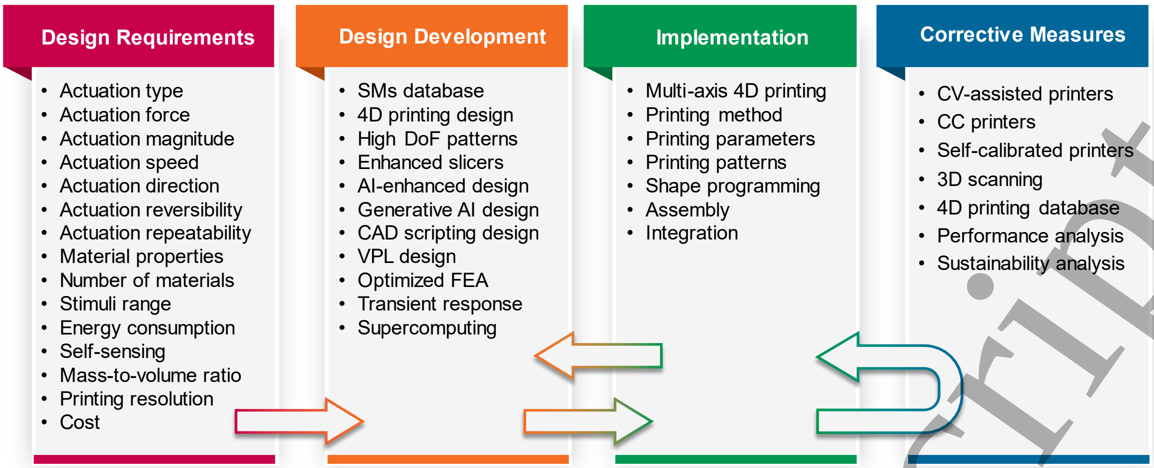

Figure 2. Future trends in pattern-controlled 4D printing.

Concluding Remarks

Pattern-controlled 4D printing offers promising advantages that keep attracting the attention of researchers. Pattern-controlled design will play an essential future role in the development of 4D printing with additional manufacturing axes. The collaboration between the industry and academia is a key factor in promoting this technology, where developing new design and simulation tools is a mutual goal for both parties.

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22 - Topology Optimization for 4D Printing

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Status

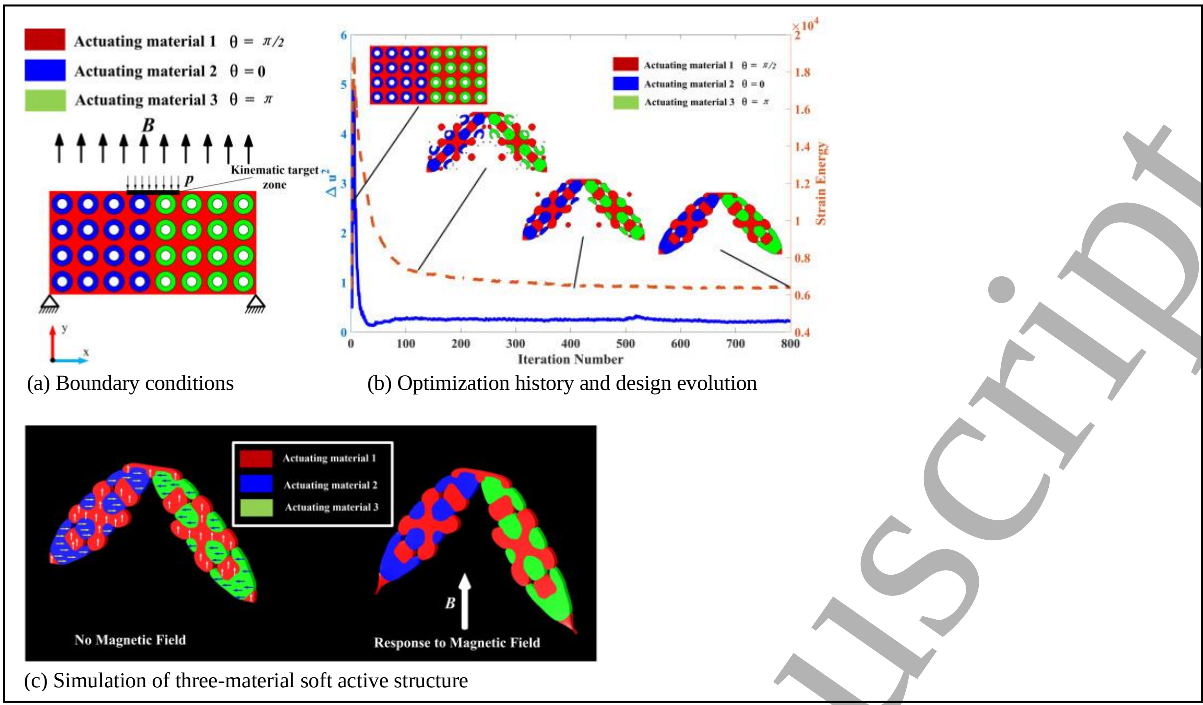
Topology optimization is a structural optimization method that has great potential to fully exploit the significant benefits provided by the increased design freedom offered by additive manufacturing or 3D printing [1-3]. 4D printing can fabricate an object that can change its color, volume, and shape under different environmental conditions [4,5], which can provide more design freedom for topology optimization. Traditionally, the purpose of topology optimization is to obtain the lightweight structure with high stiffness. By contrast, the purpose of topology optimization for 4D printing is to find the optimal distribution of component materials or building blocks that leads to desired transformations.

Current research about topology optimization for 4D printing focuses on the design of soft active structures [6-8], and multi-material structures are designed to obtain a desired shape change as multiple combinations of materials can be generated to achieve a specific goal in comparison with single material optimization [7,8]. The effectiveness of topologically optimized soft active structures was numerically and experimentally validated by Tian et al. [7]. The topological design of a three-material soft active structure presented in Figure 1 exemplifies the potential applications of current topology optimization techniques to 4D printing design. In Figure 1, the environmental stimulus is the magnetic field, θ represents the magnetization direction, ***p*** represents the loading that is distributed on the contacting area (the black area in Figure 1(a)), ***B*** represents the magnetic flux density, and Δu^2 is the least-square error with respect to the target displacement. One typical application of topology optimization to 4D printing is the design of the soft actuator for delivering delicate tasks in fragile environments, for example, food and biomedical sectors [9]. In addition, a theoretical framework of topology optimization for 4D printing was established by Garcke et al. [10].

Current and Future Challenges

Although the challenges and corresponding solutions concerning topology optimization for traditional 3D printing have been systematically discussed [3], 4D printing oriented topology optimization has not been widely studied yet. Some works used the linear elastic material model for simplicity [7,9]. However, soft active structures will generate large deformations under a certain environmental stimulus, and hence material and geometrical nonlinearities should be incorporated into topology optimization. The most challenging aspect is establishing an accurate relationship between the structural response and the corresponding environmental stimulus. If too many assumptions are made in optimization, the effects of the environmental stimulus on the structural response cannot be controlled accurately, resulting in a solution far from the actual requirements. The fabrication of topologically optimized multi-material soft active structures is challenging due to the material and geometrical complexities. The manufacturability of topologically optimized soft active structures in the literature has not been fully validated yet, and therefore most of them are conceptual designs.

Topological design of lattice structures or metamaterials for 4D printing can be regarded as the future challenge. Although 4D metamaterials with zero Poisson's ratio, shape recovery, and energy absorption features have been studied [11], topology optimization is not used for the design of 4D metamaterials. Topological design for metamaterials requires extra computational methods such as the homogenization theory to design structural configurations with periodic boundaries and repetition of representative unit cells [12]. When it comes to 4D metamaterials, a more complicated mathematical model is required.



(a) Boundary conditions (b) Optimization history and design evolution

(c) Simulation of three-material soft active structure

Figure 1. Topologically optimized three-material soft active structure with response to a magnetic field [7].

Advances in Science and Technology to Meet Challenges

The existing works regarding geometrical and material nonlinearities in [13,14] can be easily used to consider large deformations under a certain environmental stimulus for 4D printing. Feng et al. [15] conducted additive manufacturing oriented multi-material topology optimization to overcome the size limitations of the 3D printer, and this work can be employed to increase the manufacturability of the optimized multi-material soft active structures by considering the dimensional constraints of each material component. Han and Wei [16] investigated multi-material topology optimization and additive manufacturing for metamaterials, which can be extended to 4D metamaterials via smart materials. Li et al. [17] systematically carried out topology optimization of multi-material structures with programmable elastic responses under finite deformations, and this work can be modified to construct an accurate relationship between the structural response and environmental stimuli. Solutions to the aforementioned challenges were studied separately, and hence a proper combination of different fields is needed to obtain manufacturable topologies that are capable of responding to different environmental stimuli accurately or at least remaining an acceptable deviation.

Concluding Remarks

Current works mainly concentrate on the topological design of multi-material soft active structures. This research topic is still in the theoretical stage. Large deformations, manufacturability of multiple materials, and accurate relationship between structural responses and different environmental stimuli need to be incorporated into topology optimization to yield practical solutions for 4D printing. The topological design of metamaterials for 4D printing will be a promising research direction in the future.

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23 - 4D Artifacts

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Status

4D artifacts refer to a category of objects made from programmable materials with digital manufacturing technology represented by 4D printing. Beyond three-dimensional (3D) space, a 4D artifact can transform itself into another structure under the influence of external energy input, such as temperature, light, or other environmental stimuli, and therefore achieve smart functionalities such as self-deformation [1], self-healing [2], or self-assembly [3]. The 4D concept has gained considerable and significant development across various fields, such as materials science [4], medical engineering [5], and robotics [6].

In addition to functional advancements, the consideration of human and societal impact of artifacts throughout the whole 4D process [7] (i.e., 4D manufacturing, usage, and reconfiguration) has become an ongoing topic of interest to scientists, human-computer interaction researchers, and designers. With the development of universal 3D printing and personal additive manufacturing technologies, there is a growing development in the design and manufacturing technology of democratized 4D artifacts, including in the fields of 3D printing, smart materials, programmable structures, shape-changing mechanism, etc.

Digital manufacturing technologies (see Figure 1a), such as 3D printing, serve as the foundation for realizing 4D artifacts. Techniques like fused deposition modeling 3D printing (FDM3DP), direct ink writing (i.e., DIW, PP), and cured printing (i.e., SLA, SLS) are widely employed for fabricating 4D artifacts designed with computer-aided tools. Prominent examples are FDM3P thermoplastics (e.g., PLA, ABS, PCL, etc.) [8-15], which possess the advantages of low cost, high availability, excellent comprehensive performance, and reusability. Direct ink writing or jetting technology [16-17] includes extrusion modeling of semi-liquid materials and inkjet modeling of liquid ink. However, due to the properties of the liquid material, the adapted printing machine usually requires customization, and the complexity of morphogenesis is limited. Cured printing technology [18] can achieve more complex and intricate three-dimensional shapes, such as suspended and stacked with the support of the powder or liquid material itself, albeit at a relatively high cost and threshold.

According to the "smart" properties of the material (Figure 1b), which are usually reflected in the shape-change or shape-memory properties under certain external stimulus conditions, structural design (Figure 1b) is carried out through programming to allow for precise control over its transformation between different states (Figure 1c).

Currently, research projects are focused on the development of customized materials, design and fabrication tools, and post-processing methods to improve shape-changing controllability and manufacturing efficiency, and on expanding shape-shifting possibilites and embedding electronics to enhance the interactivity of 4D artifacts. Beyond these, the integration of shape-changing technologies may bring about disruptive changes in the traditional manufacturing industry (Figure 1d-f). For example, the exploration of flat production of 4D pasta [19], which can take shapes during cooking, can dramatically improve manufacturing efficiency, conserve storage and transportation space, improve cooking efficiency, and elevate the consumer experience. Moreover, further development on integrating

4D technology throughout the entire lifecycle of artifacts may present unique opportunities and challenges for sustainability.

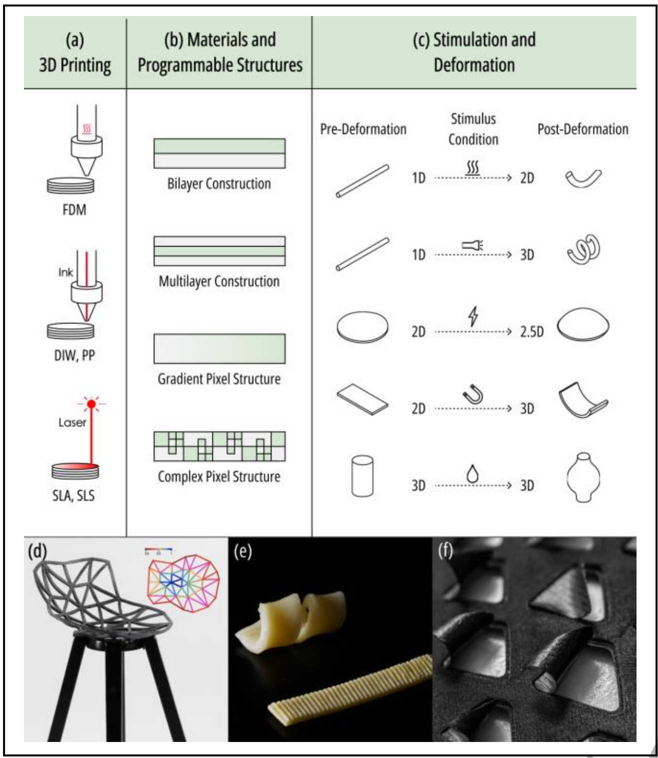

Figure 1. Schematic of the core design element of 4D artifacts including the field of (a) 3D printing, (b) smart materials, programmable structures, (c) stimulus conditions, and deformation behaviors. Examples of cutting-edge 4D artifacts include (d) 4D furniture [9], (e) shape-changing pasta [19], and (f) breathing fabrics [15].

Current and Future Challenges

To establish 4D technology as a sustainable, consumer-grade, and industrial-scale method for 4D artifacts, the challenges can be categorized across the entire life cycle of 4D artifacts into knowledge, technological, design, humanistic, and industrial challenges.

Knowledge challenges. Research into the principle of material deformation is crucial in the pursuit of 4D artifacts. When confronted with unknown deformation principles and mechanisms, researchers are often required to possess interdisciplinary knowledge integration capabilities and collaborative innovation in materials science, mechanical engineering, computer science, design, etc. Furthermore, the development of smart materials can help optimize form factors to solve practical issues, such as minimal weight, high resolution, or low cost. Consequently, the investigation of materials with unconventional functionalities has always been an enduring challenge for 4D artifacts.

Technology challenges. A significant challenge still exists in achieving precise control over high-resolution deformation based on smart materials. Aligned with broader computing trends, the form factor of 4D artifacts is moving from stationary to mobile, wearable, and even implantable. They are also evolving from one-way transformations to reversible, temporal, and even multi-state changes on demand. Besides investigating material properties, to realize this vision, increasing manufacturing precision and speed, enhancing actuation reliability and safety, and simplifying digital design and manufacturing technologies remain challenging to achieve highly accessible customized form factors and actuation methods.

Design challenges. The most substantial differences from traditional design are the form of responsive artifacts and their dynamic attributes. A crucial challenge arises when translating static forms from traditional design representations, such as sketches and prototypes, into actual 4D artifacts with temporal characteristics. Designers, in particular, will be challenged to develop 4D artifacts that are satisfying in both the form and dynamics of interaction between artifacts and people interacting with them, as well as how to predict and verify the design at a low cost in advance.

Humanistic challenges. From a humanistic perspective, the viability of 4D artifacts as a reliable and useful medium for human-object interaction is underpinned by a comprehensive understanding of the user experience when using 4D artifacts. This understanding enables us to elucidate their value, identify the domains and tasks in which they offer advantages, and facilitate their design and construction. Nonetheless, there are uncertainties and challenges to formulating theories and principles regarding the acceptability of their transit interactive behaviors to the broader public, as well as how to predict and respond to users' interactions with new 4D artifacts.

Industrial challenges. 4D artifacts may encounter challenges in terms of short-term benefits, such as increased resource demand and higher costs of recycling and reuse, but the ultimate goal of the 4D concept is to establish a novel ecological industrial chain that spans the entire lifecycle of 4D artifacts to achieve long-term sustainability. This revolution seeks to reshape established consumption habits by realizing multiple functions within a single 4D artifact [20], such as one 4D personal computer displacing a number of mobile phones, watches, computers, laptops, etc. Therefore, the primary challenges for 4D artifacts lie in ensuring their "lifelong" deformation functionality, where a new industrial standard for 4D artifacts should be established instead of adhering to a consumer-level definition.

Advances in Science and Technology to Meet Challenges

To successfully address the current and future challenges of 4D artifacts, it is necessary to take a holistic view that considers both the vertical life-cycle chain of 4D artifacts and the horizontal relationships between humans and the artifacts (Figure 2).

In tackling the challenges within the vertical life-cycle chain of 4D artifacts, the rapid and lightweight development of additive manufacturing concepts can be important. Leveraging cutting-edge materials science and nanotechnology, coupled with advanced simulation and modeling tools, holds the potential to deepen our comprehension of materials' time-dependent properties and grant us greater control over them. This, in turn, facilitates dynamic manufacturing processes and on-demand deformations. Additionally, the enhancement of forward design capabilities and inverse simulation computing will accelerate the development of 4D manufacturing technology, enabling engineers to predict and optimize the behavior of 4D printed objects, thereby reducing resource waste. Furthermore, the development of innovative 4D artifacts, under the guidance of new user experience requirements, is imperative for top-down guidelines for improving the vertical life cycle chain.

Confronted with the challenges in the horizontal relationship between humans and artifacts, optimizing the end-user interaction experience is critical. The integration of intelligent control systems and human-machine interfaces will enhance the practicality and usability of 4D artifacts. At the same time, addressing the safety and ethical considerations surrounding 4D artifacts is integral to its democratization. Sustainability concerns also must factor into material selection, production efficiency, and product lifespan to mitigate environmental impact and ensure the sustainable development of 4D technology.

In summary, 4D artifacts ultimately return to humans, and their development holds the promise of offering new solutions to the issues impacting humans, society, and the environment.

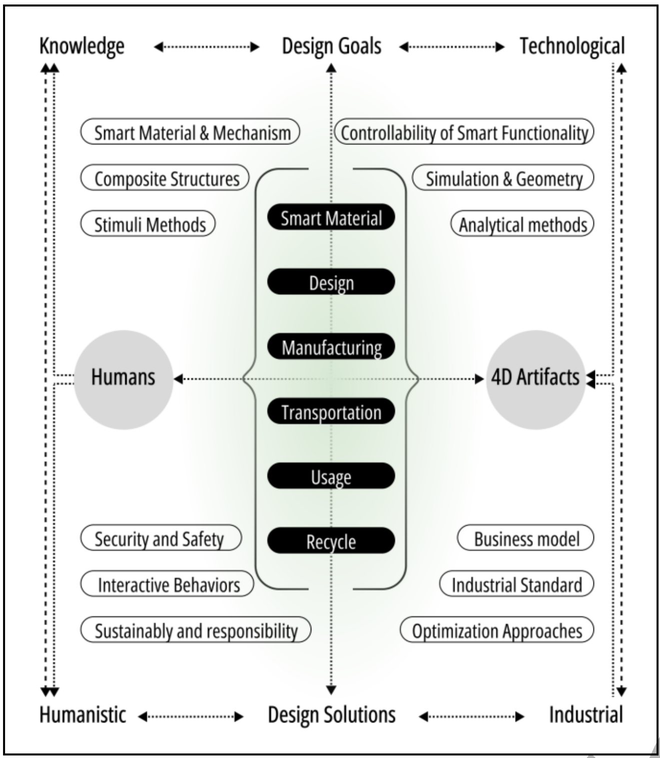

Figure 1. The challenges and opportunities of 4D artifacts in a holistic view of the vertical life-cycle chain of 4D artifacts and the horizontal relationship between humans and artifacts.

Concluding Remarks

Despite the fact that 4D printing remains in its early stages and there is still a long way to go before its commercialization and widespread application, as an emerging additive manufacturing technology, 4D printing is promising to become an efficient, sustainable, and personalized manufacturing method. At the same time, 4D artifacts have the potential to provide end users with adaptive, augmented, and versatile interactive experiences towards enormous applications in intelligent interactive terminal products, human-machine hybrid interaction, biomedical, and other fields.

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24 – Multi-Axis 4D Printing

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Status

Three-dimensional (3D) printing has experienced great advancements, especially in the area of fused deposition modeling (FDM) printers, where many manufacturers have developed advanced types of FDM 3D printers, robotic-arm-based printers, and aerial 3D printers [1]. Most researchers use FDM 3D printers that support moving the printing head or bed along the *x*-, *y*-, and *z*-axes. However, the main configuration that is used by researchers is 2.5-axis 3D printing due to the incomplete control over the deposition along the *z*-axis. In this approach, the model is sliced into parallel planar layers that are then printed layer-by-layer using a specific increment along the *z*-axis. Nevertheless, researchers were able to tackle this issue by developing advanced slicing methods that support 3-axis printing [2], as well as 3D printers that support multi-axis 3D printing. This allowed achieving 3D printing of structures using multi-axis configurations, including 4-axis [3], 5-axis [4], 6-axis [5], 7-axis [6], and 8-axis [7]. These continuous, or simultaneous, multi-axis printing approaches opened the doors for sub-categories based on these approaches, which are known as positional, or indexed, configurations. Some examples include 3 + 1-axis [8], 3 + 2-axis [9,10], and 3 + 3-axis [11].

Four-dimensional (4D) printing relies on the shape programming of structures that were 3D-printed from smart materials. This process allows these structures to change their shape, color, or function over time, which adds a fourth dimension to the 3D printing process [12]. For instance, when using FDM, the shape-programming approach is mainly carried out during the printing process along the printed lines, which is also performed using a 2.5-axis configuration [13]. Therefore, multi-axis 4D printing offers great potential for complex multi-axis shape programming, where the strain can be induced along additional printing axes. So far, multi-axis 4D printing has witnessed limited progress, where researchers have only demonstrated 6-axis 4D printing [14] and 6 + 1-axis 4D printing [15]. However, it seems that the terminology of such a process is not well-understood, where some researchers refer to this process as 5D or 6D printing [16]. These definitions might be subject to misinterpretation since there is no additional dimension added to the printed structure when using additional printing axes. For instance, some manufacturers may refer to their printers as 5D printers, but this is a short term that refers to the degrees-of-freedom (DoF), which is different from the time-dependent behavior attributed to the fourth dimension. Therefore, we believe that there is a high need for standard terminology in multi-axis 3D and 4D printing due to the rapid development in these areas. Figure 1 presents potential advancements in multi-axis 4D printing.

Current and Future Challenges

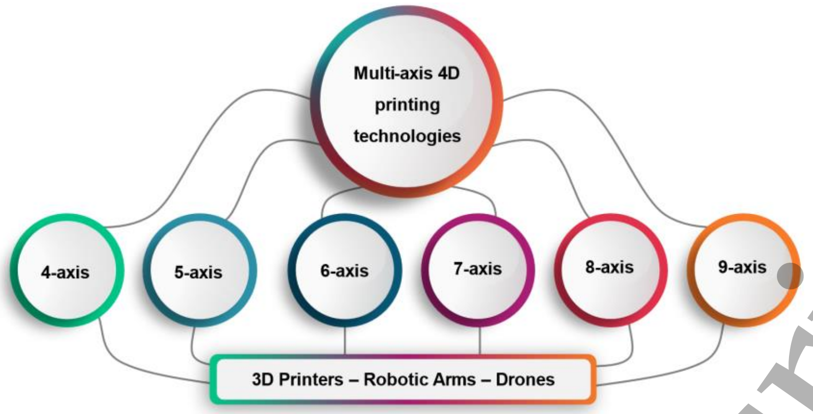

Figure 1. Main potential multi-axis 4D printing technologies.

Multi-axis 3D/4D printing strategies that utilize a high number of axes, such as 5-axis 3D/4D printing, offer several structural advantages compared to 2.5-axis 3D/4D printing. This is due to the fact that the former consumes less printing material while offering the ability to produce stronger structures compared to the latter. However, increasing the number of axes used in the printing process comes with several challenges that hinder progress in this area. For instance, the complexity and cost of 3D printers and systems that support such printing methods increase significantly as the number of axes increases. In many cases, the additional printing axes are realized by adding rotational movements to the printing head or printing bed, by using a robotic arm, or by a combination of all these possibilities. This also leads to an increase in the complexity of slicing such designs, where standard slicers that support 2.5-axis 3D printing are not suitable for these approaches. This complexity is caused by the challenges associated with planning the printing path in 3D space. Moreover, such a printing process causes interference between the printing paths when printing non-uniform objects. This also leads to challenges when selecting the orientation of the printed object, as well as the orientation of the printing head along each printing path to avoid colliding with the printed part. Furthermore, multi-axis 3D and 4D printing require a long time and high energy consumption to print objects due to the constant change in orientation of the printing head and printing bed.

These challenges become more evident when developing structures for multi-axis 4D printing. The design requirements of 4D printing add more challenges to the whole printing process in terms of hardware implementation, slicing, path planning, modeling, and simulation. For example, FDM-based strain-induced shape programming faces several challenges when using this approach. The levels of induced strain in each layer and each part of the printed structure need to be precisely controlled to achieve the desired shape change after activating the structure. This means that the printing parameters need to be carefully designed to meet the additional requirements of multi-axis 4D printing. For instance, the reported modeling and simulation approaches to 4D-printed structures only consider planar layers to estimate the bending angle and shape-morphing of such structures [17]. Therefore, there is a need for advanced modeling and simulation approaches that can handle the complexity of multi-directional induced strain.

Advances in Science and Technology to Meet Challenges

Currently, there is a lack of development in the area of multi-axis 4D printing since this technology is still in its early stages, where the development is mainly focused on 2.5-axis 4D printing. Therefore, most research is dedicated to tackling the challenges associated with multi-axis 3D printing without considering the requirements of 4D printing. Significant research has been undertaken to address the issues associated with slicing designs for multi-axis 3D printing, such as feature-based slicing, adaptive slicing, multidirectional slicing, and conformal slicing. Moreover, path planning has been one of the main focus areas in multi-axis 3D printing, which is performed after the slicing process to generate the printing toolpaths. This process can be realized using several approaches, such as online and offline collision detection and avoidance, contour-parallel printing path generation, continuous spatial path generation, gradient infill generation, as well as multi-robot path planning.

It is expected that multi-axis 4D printing will experience great advancements in the near future. This is caused by the rapid development of slicing and path-planning methods, which offer a high level of control over the paths of the induced strain. In addition, the current trend of increased quality in multi-axis 3D printers and robots indicates that 4D printing technology will have the required tools to meet the complexity of shape programming in 3D space. One of the promising areas that might see great investigation in the future is the use of teams of robotic arms in the 4D printing process, which offers great advantages in terms of increasing the DoF of the printing process. This will also open doors to creating large-scale 4D-printed structures that are not limited to the size of 3D printers. Moreover, 4D printing might benefit from the great flexibility of aerial robotics, where a drone or a swarm of drones can 4D-print structures at remote locations. Meanwhile, these advancements will also require crucial support from other technologies, such as artificial intelligence (AI), which can be used in several 4D printing areas, such as AI-enhanced design, generative AI design, and AI-based slicing and path planning. Lastly, the combined advantages of AI and finite element analysis (FEA) can be used to model the complex actuation performance of multi-axis 4D-printed structures, which may require using supercomputers to handle such complexity. A summary of the future trends in multi-axis 4D printing is presented in Figure 2.

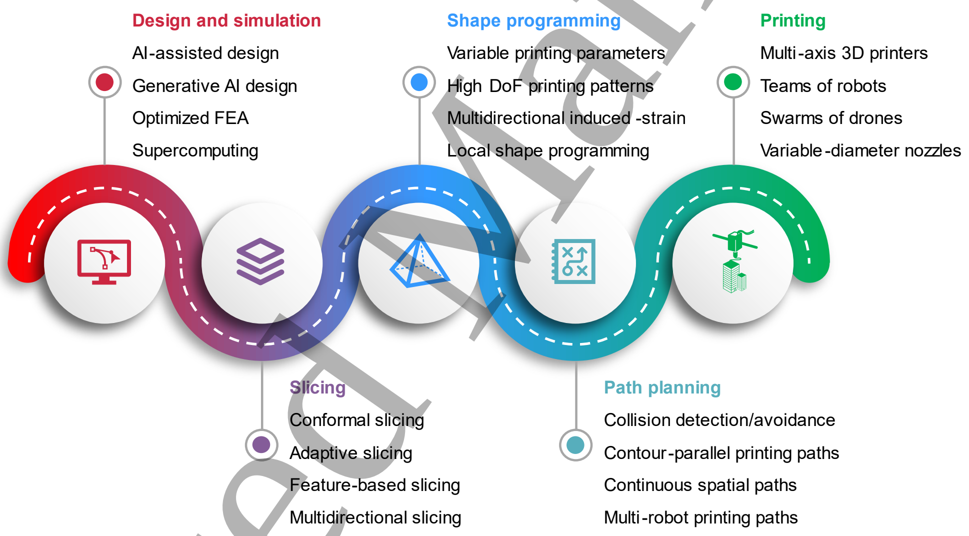

Figure 2. Future trends in multi-axis 4D printing.

Concluding Remarks

Multi-axis 4D printing offers promising advantages that can broaden the horizons of 4D printing. This technology will play a key role in the future development of 4D printing. There is a high need for collaboration between academia and industry to develop the tools required to advance this technology. Moreover, the terminology used in this research area requires further standardization and careful consideration. The authors propose the terminology of *n*-axis 3D/4D printing to differentiate products fabricated using a range of printers with modified axes, either as conventional 3D or time-responsive 4D constructs.

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