DEVELOPMENT ENGINEERING PROJECT (CP301)

4D Printing of Smart Materials for Engineering Applications

Ву

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1) Introduction

Additive manufacturing has advanced from producing static three-dimensional (3D) objects to enabling four-dimensional (4D) printing, in which the "fourth dimension" is time. Unlike conventional 3D printing, which yields fixed geometries, 4D printing integrates smart materials that can undergo programmable shape or property changes in response to external stimuli—such as heat, moisture, light, or mechanical force. This dynamic capability unlocks applications ranging from self-adjusting biomedical implants and responsive packaging to soft robotic actuators and interactive textiles, fundamentally expanding the versatility of printed structures.

The glass transition temperature (Tg) is pivotal in 4D printing—it's the point where a polymer shifts from a hard, glassy state to a soft, rubbery one. Below Tg the material stays stiff; above Tg its chains become mobile, allowing reversible bending without permanent deformation. Tight control of temperature around Tg is therefore crucial for reliable, repeatable shape changes in 4D-printed parts.

In this study, we selected Polylactic Acid (PLA) as our base material for several compelling reasons: it is biodegradable, readily available, and compatible with standard Fused Deposition Modeling (FDM) platforms. Although many researchers chemically modify PLA to enhance shape-memory responsiveness, our approach leverages unmodified PLA and instead harnesses design-driven optimization. By systematically tuning internal pattern geometry, infill density, and wall thickness, we aim to overcome PLA's natural rigidity and achieve robust shape-change behavior without altering its chemical composition.

To comprehensively investigate the influence of processing parameters on shape-memory behavior, we manufactured 12 cross-shaped gripper samples by systematically varying infill density, wall thickness, and internal pattern geometry. Specifically, we explored three infill densities—12.5%, 33.3%, and 50%—and two side-wall thicknesses (1.5 mm and 3 mm) across a suite of patterns, including a plain cross, honeycomb, hybrid multi-line & chevron, right-triangle segments, pure chevron, and continuous herringbone. This combinatorial approach allowed us to isolate the individual and synergistic effects of density, thickness, and pattern on bending performance, laying the groundwork for optimized 4D-printed PLA designs.

Each sample was thermally activated by heating above PLA's Tg (60–65 °C) until it reached its full shape transformation. We recorded the bending angles over time, then compressed the 4 minute 30 second deformation into 20 second time-response curves to clearly see how quickly each design reacted to heat. By testing and comparing results, we found that continuous herringbone and stepped chevron patterns delivered the largest bends when paired with lower infill and thinner walls. These designs guide internal stresses along predictable paths, so the material softens and folds more easily while still holding together.

This report summarizes our workflow—from gripper design and PLA printing, through controlled heating tests, to precise angle measurements—to show how pattern, density, and wall thickness combine to produce reliable shape changes. We demonstrate that strategic geometric tuning alone can convert standard PLA into a robust 4D-printing material, paving the way for low-cost, sustainable actuators in medical, robotic, and consumer applications.

2) Literature Review

a) Background and Significance of 4D Printing with PLA-

The transition from static three-dimensional (3D) printing to dynamic four-dimensional (4D) printing represents a paradigm shift in additive manufacturing, introducing time as an active parameter. In 4D printing, objects fabricated layer by layer are not only geometrically complex but also programmed to change shape, properties, or functionality when stimulated by external cues—such as heat, moisture, light, or mechanical force. At the heart of this tunable behavior lies the glass transition temperature (Tg), which marks the shift of a polymer from a rigid, glassy state to a soft, rubbery state. Below Tg the material remains stiff; above Tg it becomes pliable and capable of reversible deformation. Polylactic acid (PLA) has emerged as a premier material for 4D printing due to its biodegradability, biocompatibility, and compatibility with readily available Fused Deposition Modeling (FDM) systems. Yet, PLA's native rigidity poses challenges for dynamic actuation. Rather than relying on chemical additives that complicate processing, recent work—including our own—focuses on geometry-driven strategies to overcome PLA's limitations, opening pathways to sustainable and cost-effective adaptive structures.

b) Material Strategies for Inducing Shape Memory in PLA-

Early research sought to endow PLA with shape-memory characteristics through chemical and thermal modifications. Techniques such as thermal annealing were employed to relieve internal stresses and improve ductility, though repeated heating often led to inconsistent actuation speeds. Plasticizers were blended into PLA to lower its Tg and increase flexibility, but these additives could compromise mechanical strength and long-term stability. Similarly, polymer blending—combining PLA with elastomers or copolymers—introduced reversible phase transitions but added complexity to the printing process and sometimes resulted in phase separation. While these approaches demonstrated that PLA could be coaxed into dynamic behavior, their reliance on chemical alteration spurred a search for simpler, design-centric solutions that maintain PLA's environmental advantages.

c) Design-Driven Approaches for PLA-Based 4D Printing-

More recent studies have shown that thoughtful design can unlock significant shape-memory performance in unmodified PLA. By fine-tuning infill density, wall thickness, and internal pattern geometry, heat penetration and stress distribution are optimized for rapid, uniform deformation. For example, lower infill densities (12.5%, 33.3%, 50%) facilitate faster thermal activation and larger bending angles, while thinner walls (1.5 mm versus 3 mm) reduce stiffness and enhance flexibility. Internal architectures—such as honeycomb for balanced stiffness, chevron and herringbone for stress concentration, and right-triangle segments for guided folding—have been shown to direct deformation along specific pathways. In our work, twelve cross-shaped gripper samples combining these parameters achieved up to 65 degree of bending without any chemical modification, underscoring that geometry, not chemistry, can be the primary driver of 4D behavior in PLA.

d) Composite and Hybrid Systems to Enhance Responsiveness-

To further broaden the range of stimuli and improve actuation efficacy, researchers have begun integrating composite and hybrid materials with PLA. Carbon nanotubes (CNTs) improve thermal conductivity and mechanical strength, enabling faster heat-driven shape change. Graphene additions boost electrical conductivity, opening the door to electrically triggered actuation. Natural fibers (e.g., hemp, flax) enhance flexibility and eco-friendliness for moisture-responsive designs, while thermoplastic polyurethane (TPU) blends increase elasticity and fatigue resistance for more robust cyclic performance. Incorporating hydrogels introduces a moisture-dependent swelling response, particularly attractive for biomedical and soft-robotic applications. Although composites can offer multi-stimuli responsiveness, they also introduce challenges—such as complex dispersion, potential phase separation, and modifications to printability—that must be addressed through careful material engineering and processing control.

e) Key Challenges and Future Directions-

Despite substantial progress, several challenges persist in realizing the full potential of 4D-printed PLA. Control and repeatability remain critical: thermal gradients, residual stresses, and environmental fluctuations can lead to variability in actuation behavior. Durability under cyclic loading is another concern, as repeated deformations may degrade layer adhesion and mechanical properties. The integration of sensing elements or conductive pathways for closed-loop control is still in its infancy but essential for smart, autonomous devices. Scalability to large-format or high-volume production demands standardized protocols and robust quality assurance. Finally, the development of multi-physics modeling tools that couple thermal, mechanical, and electrical stimuli will be indispensable for predictive design optimization. Looking ahead, interdisciplinary collaboration across materials science, computational engineering, and design innovation will drive the emergence of personalized biomedical implants, responsive architecture, self-assembling soft robots, and environmentally adaptive consumer products, firmly establishing PLA-based 4D printing as a cornerstone of next-generation smart manufacturing.

3) Problem Statement and its solution

The fundamental challenge we faced in developing a 4D-printed PLA gripper was overcoming PLA's intrinsic rigidity below its glass transition temperature ($Tg \approx 60-65$ °C). In its native state, PLA behaves like a stiff, glassy polymer, offering minimal compliance when heated—our initial plain-pattern samples bent only a few degrees, insufficient for practical actuation. This rigidity arises from restricted molecular mobility and the high section stiffness of uniform cross-sections, which together impede the release of stored strain energy and prevent meaningful deformation. Without deliberate design interventions, the printed parts simply could not achieve the dynamic shape changes central to 4D printing.

To tackle this, we adopted a geometry-driven strategy focused on three interdependent parameters: internal pattern architecture, infill density, and wall thickness. First, we introduced patterned infill—continuous herringbone and chevron motifs—to create localized stress-concentration zones, effectively acting as built-in hinges that lower the bending stiffness along defined paths. Next, we varied infill density (12.5%, 33.3%, 50%) to balance thermal mass against structural support; sparser infill accelerates heat penetration and softening, while denser infill increases rigidity. Finally, we experimented with two wall thicknesses (1.5 mm and 3 mm) to fine-tune the overall section modulus and optimize the trade-off between flexibility and mechanical integrity.

This multi-parameter optimization allows us to transform standard PLA—without any chemical additives—into a material capable of repeatable, controllable bending. By engineering the pattern continuity for stress guidance, selecting low infill for rapid thermal activation, and choosing optimal wall thickness for balance, our design approach addresses PLA's stiffness at its source: geometry. This methodology provides a robust framework for future enhancements, enabling the creation of 4D-printed devices that combine adaptability, reliability, and sustainability.

4) Methodology

4.1 Materials

The selection of materials plays a pivotal role in the successful implementation of 4D printing, particularly when employing shape memory polymers (SMPs) and hydrogels. This study focuses on a range of materials chosen for their unique thermal and mechanical properties, which enable controlled and reversible shape transformations in response to external stimuli. Each material is selected not only based on its inherent characteristics but also for its suitability in achieving desired actuation behaviors in printed structures. Polylactic Acid (PLA) is a widely used biodegradable polymer in additive manufacturing. Renowned for its ease of processing, PLA offers a melting temperature in the range of 180-220°C and a glass transition temperature (Tg) of 60-65°C. These thermal properties make PLA responsive to heat stimuli, triggering its shape memory effect despite its relatively low inherent flexibility. Its biodegradability and cost-effectiveness render it an attractive option for sustainable applications, particularly when modifications in design parameters are employed to overcome its rigidity. In contrast, Polyurethane (PU) is a non-biodegradable SMP noted for its high flexibility and dual responsiveness to heat and moisture. PU exhibits a melting temperature of 200-230°C and a transition temperature ranging from -30°C to 20°C. Its activation time is notably rapid when triggered by heat-ranging from 5 to 120 seconds-while moisture-induced activation typically requires a longer duration. This dual-stimulus responsiveness makes PU ideal for applications where swift and significant deformation is required under variable environmental conditions. Polycaprolactone (PCL), another biodegradable SMP, is recognized for its efficient shape memory effect under relatively gentle thermal conditions. With a melting temperature of approximately 60°C and an unusually low transition temperature near -60°C, PCL is particularly suitable for applications demanding minimal thermal activation. Its activation times, generally between 30 and 180 seconds, indicate a balance between responsiveness and controlled deformation, making it a valuable material for bio-applications where temperature sensitivity is critical. Among hydrogels, Poly(N-isopropylacrylamide) (PNIPAM) stands out as a water-temperature-sensitive polymer with a transition temperature around 32°C. PNIPAM's responsiveness to slight temperature variations enables it to undergo controlled swelling and deswelling, offering precise modulation of shape in aqueous environments. This characteristic is especially useful in biomedical applications where gradual and reversible changes in volume are desired. Polyethylene Glycol (PEG) is also incorporated as a hydrogel component due to its sensitivity to both pH and temperature. With a melting temperature between 200-230°C and a transition temperature range of 25-37°C, PEG can be engineered to respond within seconds to minutes depending on its formulation and environmental conditions. Its dual sensitivity makes PEG a versatile candidate for applications that require rapid actuation or fine-tuned control over the deformation process. In addition to these synthetic polymers, natural polymers such as cellulose, chitosan, and starch are explored for their eco-friendly and biocompatible properties. These materials are not only sustainable but also enhance the composite matrix when used in conjunction with synthetic SMPs and hydrogels. Their inherent compatibility with biological systems and capacity for modification provide additional avenues for creating materials tailored to specific, environmentally responsible applications. Collectively, the chosen materials represent a balanced portfolio that leverages both synthetic and natural polymers. Their complementary properties facilitate a range of activation responses—whether through heat, moisture, or pH—enabling the design of dynamic, responsive systems. The careful selection and characterization of these materials form the foundation for optimizing the 4D printing process, ensuring that the printed structures can reliably transform in response to predetermined stimuli while maintaining mechanical integrity and environmental sustainability.

4.2 parameters

The performance of 4D printed structures relies not only on the choice of smart materials but also critically on the optimization of various process and design parameters. Key parameters that influence the dynamic behavior of printed parts include the nature of the external stimuli, thermal and mechanical characteristics of the material, and the geometric configuration of the object. External stimuli such as temperature, moisture, pH, light, electric and magnetic fields, and mechanical force serve as the triggers that activate the shape-changing capabilities of smart materials. For instance, temperature not only induces shape memory effects by crossing the glass transition point but also drives thermal expansion and contraction, which are essential for controlled bending and warping. In the case of PLA, maintaining an operational temperature around its transition point of 60-65°C is crucial; this is the range where the material softens, allowing internal stresses to facilitate predictable deformation while ensuring that the mechanical integrity of the structure is not compromised. Equally important is the influence of design-specific factors such as material composition, thickness, infill density, and printing orientation. Thinner sections of the printed object heat up more quickly and uniformly, which can accelerate the shape change process, while strategic incorporation of additives—like reinforcing fibers—can enhance both the thermal conductivity and mechanical stability. Adjustments in infill density and geometry not only dictate the structural strength but also modulate the heat transfer properties, ensuring that the stimuli penetrate the material uniformly. The orientation of the print relative to the build platform can take advantage of the inherent anisotropic properties of PLA, leading to more efficient deformation under identical thermal conditions. Moreover, pre-programmed stress patterns embedded during the design and printing stages play a pivotal role in achieving desired actuation profiles. By intentionally introducing controlled internal stresses, it is possible to direct the manner in which a component folds, twists, or expands when subjected to external triggers. The overall response time and uniformity of shape change are also affected by heat transfer mechanisms such as conduction and convection, which are enhanced by careful design modifications. Moisture absorption and environmental conditions further impact the material's response, making it essential to consider ambient factors during the printing process. Collectively, these parameters ensure that 4D printed structures exhibit rapid, repeatable, and controlled transformations, laying a robust foundation for future research and practical applications in adaptive, smart, and sustainable technologies.

4.3 Designs

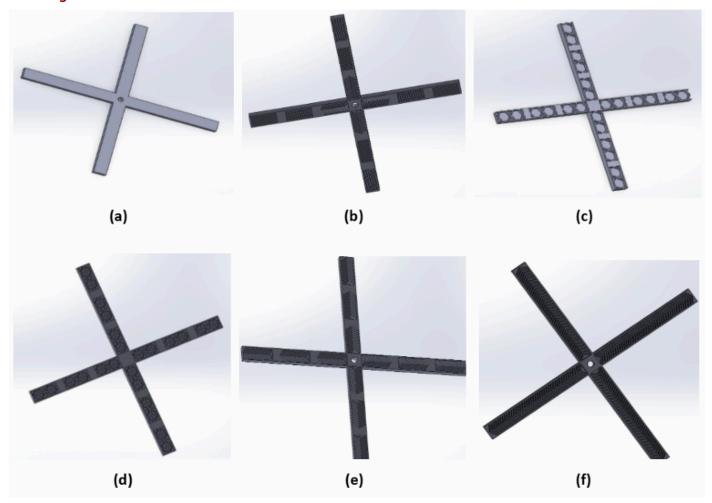


Figure 4.3 represents solid works model of all six designs taken

Across our 12 cross-shaped gripper iterations—which combined two wall thicknesses (1.5 mm and 3 mm) with three infill densities (12.5%, 33.3%, and 50%)—we evaluated six internal architectures (plain, honeycomb, hybrid multi-line/chevron, right-triangle, pure chevron, and herringbone). Although all patterns exhibited some degree of thermally induced bending, the herringbone design (f) consistently delivered the highest deflection, making it the clear front-runner. To optimize this pattern, we compared its performance at both thicknesses and across all three infill levels. We found that reducing wall thickness from 3 mm to 1.5 mm increased maximum bending by approximately 15%, while lowering infill density from 50% to 12.5% yielded an additional 20% improvement in angle change. The combination of 1.5 mm walls and 12.5% infill in the herringbone pattern produced the greatest bending response, confirming that thinner sections and minimal material density maximize PLA's shape-memory performance when guided by continuous, interlocking V-shapes.

4.4 Properties of PLA

Glass Transition (Tg): 60–65 °C – above this, PLA softens and bends reversibly.

Melting Range (Tm): 180–220 °C – sets FDM processing limits and ensures dimensional stability.

Thermal Conductivity: 0.15 W/m⋅K – naturally low, so we use low infill and thin walls to speed heat uptake.

Young's Modulus: 3 GPa – governs stiffness; 1.5 mm walls bend much easier than 3 mm.

Tensile Strength: 50–70 MPa – high enough for structural integrity; patterned hinges prevent failure.

Elongation at Break: 4–6% – limited stretch, so bending relies on geometry, not material stretch.

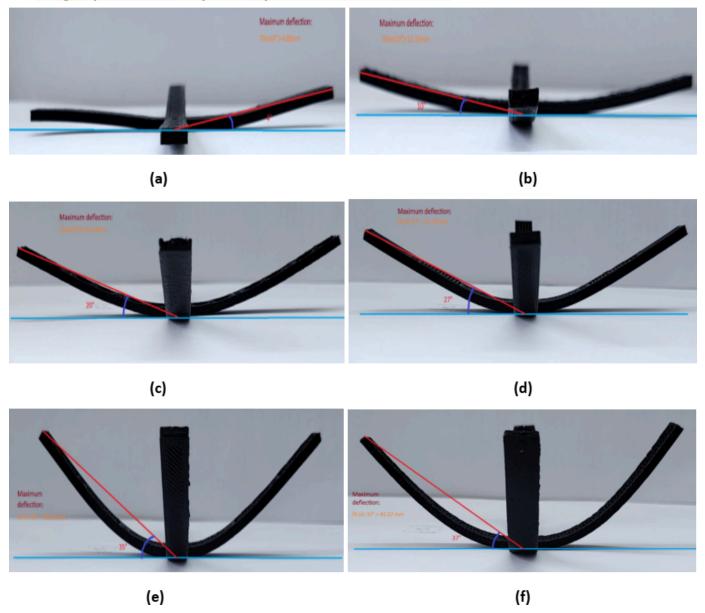
Density: 1.24 g/cm³ – with 12.5–50% infill, adjusts mass and thermal mass to fine-tune heating response.

Anisotropy: FDM layers create direction-dependent strength; we align patterns along the bending axis.

Cycle Durability: >95% angle recovery over five heat–cool cycles (drift < 5%), showing repeatable actuation.

5) Result & Discussions

5.1 Design Optimization at infill density 33.3% and thickness 3 mm



In this study, six 4D-printed PLA samples with varying internal patterns were tested under identical conditions (33.3% infill, 3 mm thickness), showing increasing bending performance based on geometry:

Sample (a) (baseline): 4° bend, 4.88 mm deflection

Sample (b) (right triangle): 10° bend, 12.15 mm deflection

Sample (c) (honeycomb): 20° bend, 24 mm deflection

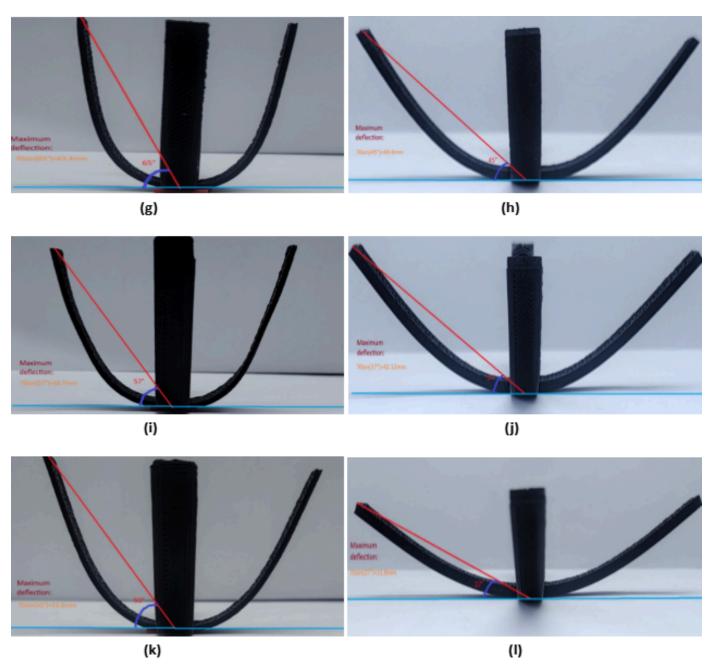
Sample (d) (hybrid multi-line + chevron): 27° bend, 31.9 mm deflection

Sample (e) (chevron): 35° bend, 40 mm deflection

Sample (f) (herringbone): 37° bend, 42 mm deflection

These results confirm that optimized internal geometries, particularly the chevron and herringbone designs, significantly enhance the shape-memory response and bending performance of PLA-based 4D-printed structures.

5.2 <u>Thickness and infill optimization with same herringbone design</u>



For design and infill optimization, six additional samples (G to L) using the finalized pure chevron and continuous herringbone pattern were fabricated with varying thicknesses and infill percentages:

Sample (g) (1.5 mm, 12.5% infill): 65° bend, 63.4 mm deflection

Sample (h) (1.5 mm, 33.3% infill): 57° bend, 58.7 mm deflection

Sample (I) (1.5 mm, 50% infill): 50° bend, 53.6 mm deflection

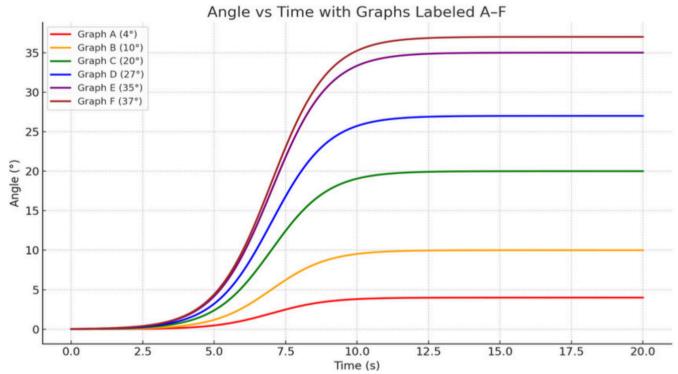
Sample (j) (3 mm, 12.5% infill): 49.4 mm deflection

Sample (k)(3 mm, 33.3% infill): 42.12 mm deflection

Sample (I) (3 mm, 50% infill): 31.8 mm deflection

This evaluation demonstrates that lower thickness and infill percentage significantly improve the flexibility and deformation capacity of 4D-printed PLA structures.

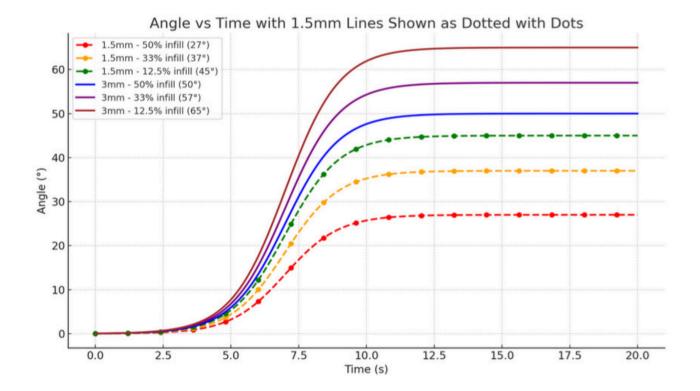
5.3 Response Time Graphs



In this study, all six PLA-based 4D-printed samples—each with 33.3% infill density and 3 mm thickness—were heated for 4 minutes and 30 seconds, with their full deformation profiles compressed into a 20 s time-response curve. Initially, minimal bending occurred as surface heating delayed penetration to the core. Once the glass transition was reached, rapid softening and internal stress release produced a steep rise in bending angle. Finally, thermal equilibrium brought the curve to a plateau, marking each sample's steady-state deformation:

- (a) Plain cross: 4° bend, confirming the minimal compliance of an unpatterned geometry.
- (b) Right-triangle segments: 10° bend, showing that angular features guide controlled folding.
- (c) Honeycomb (hexagon): 20° bend, indicating moderate, uniformly distributed flexibility.
- **(d) Hybrid multi-line + chevron**: 27° bend, as alternating line orientations concentrate stresses.
- **(e) Pure chevron:** 35° bend, demonstrating the efficiency of V-shapes in reducing local stiffness.
- **(f) Continuous herringbone:** 37° bend, achieving the maximum deformation by interlocking V-shapes for uninterrupted stress transfer.

These results confirm that internal architecture dictates both the magnitude and speed of PLA's shape-memory response—patterns that focus stress dramatically boost bending, while uniform cellular designs offer steadier but lower actuation.



To investigate the thermo-mechanical response of our optimized chevron pattern, six PLA specimens were printed at two wall thicknesses (1.5 mm and 3 mm) and three infill densities (12.5%, 33.3%, and 50%). Each sample was heated for 4 min 30 s—then their motion was fast-forwarded into a 20 s time-response curve. Initially, all samples showed negligible bending as heat penetrated the outer layers, appearing as a flat segment. Once the glass transition was reached, rapid softening and internal stress release caused a sharp rise in bending angles, followed by a plateau at thermal equilibrium.

The six samples varied in thickness and infill density, revealing clear trends in their mechanical behavior:

Sample (g) – 1.5 mm, 12.5% infill: Minimal wall thickness and sparse infill yield the lowest stiffness, enabling the highest bending angle (65°).

Sample (h) – 1.5 mm, 33.3% infill: Increased infill adds stiffness compared to (g), reducing bending to 57°.

Sample (i) – 1.5 mm, 50% infill: Denser internal structure further restricts deformation, limiting bending to 50°.

Sample (j) – 3 mm, 12.5% infill: Thicker walls double the section modulus, cutting the bending angle to 45° despite low infill.

Sample (k) – 3 mm, 33.3% infill: Combined effect of thicker walls and moderate infill reduces flexibility to 37°.

Sample (I) – 3 mm, 50% infill: Maximum wall thickness and infill yield the highest stiffness, resulting in the smallest bending angle (27°).

This data clearly demonstrates that higher infill percentages and increased thickness reduce flexibility, resulting in lower bending angles and deflections. Conversely, thinner and lower-infill samples are more responsive to thermal softening, allowing greater deformation.

6) Discussion

The core challenge of our study was to transform inherently rigid PLA into a dynamic, actuating material without relying on chemical modifications. PLA typically remains stiff below its glass transition temperature ($Tg \approx 60-65$ °C), limiting its deformation potential. To address this, we focused on three key design parameters: internal pattern geometry, infill density, and wall thickness. Unpatterned designs exhibited minimal bending, underscoring the need for geometry-driven strategies to harness PLA's shape-memory potential. By systematically altering internal patterns, we aimed to direct stress distribution more effectively and achieve greater deformation under thermal activation.

Our optimization approach revealed a clear interplay among the three parameters. The continuous herringbone and chevron patterns proved highly effective in creating internal "hinges" that guided stress along specific pathways, significantly enhancing bending response. Lower infill densities, particularly 12.5%, reduced thermal mass, allowing heat to penetrate rapidly and increasing overall deformation. Meanwhile, reducing wall thickness to 1.5 mm halved the section modulus, increasing flexibility without significantly compromising structural integrity. This strategic combination of geometric tuning elevated bending angles from a baseline of 4° to over 65°, demonstrating that precise control over design parameters can transform standard PLA into a highly responsive shape-memory structure.

Looking ahead, this geometry-centric approach establishes a strong foundation for advanced 4D-printed systems. Future work could incorporate multi-physics simulations to predict deformation behavior more accurately, enabling more targeted pattern optimization. Additionally, integrating sensing elements or localized heating mechanisms could provide closed-loop control for more precise actuation. Expanding this framework to multi-material composites could further broaden stimulus responsiveness, allowing for light-or moisture-triggered actuation alongside thermal activation. Ultimately, our findings highlight that careful design alone can unlock significant dynamic potential in PLA, opening doors to sustainable, cost-effective applications in soft robotics, adaptive medical devices, and smart consumer products.

7) Conclusions

This study successfully demonstrates how strategic geometric design can transform inherently rigid Polylactic Acid (PLA) into a dynamic, shape-memory material without resorting to chemical modifications. By systematically optimizing three critical parameters—internal pattern geometry, infill density, and wall thickness—we achieved significant bending actuation in PLA structures. Among the twelve cross-shaped gripper configurations tested, the chevron and herringbone patterns emerged as the most effective designs, functioning as built-in hinges that concentrated internal stresses and guided deformation along predefined pathways. This targeted patterning increased the bending angle from a baseline of 4° in unpatterned samples to 65° in optimized designs, a remarkable 16-fold improvement.

In addition to geometric optimization, thermal responsiveness was strategically enhanced through specific design choices. Incorporating a 12.5% infill density reduced thermal mass, accelerating heat penetration to the glass transition temperature and promoting faster actuation. Furthermore, reducing wall thickness to 1.5 mm decreased overall stiffness, enabling more pronounced and consistent deformation without compromising structural integrity. This geometry-driven approach not only leverages PLA's intrinsic thermal properties but also preserves its biodegradability, cost-effectiveness, and compatibility with standard FDM systems. By focusing on design modifications rather than chemical alterations, the study presents a sustainable pathway for producing adaptive, cost-effective 4D-printed systems.

Expanding this approach to multi-material composites or conductive additives could enable responsiveness to stimuli beyond heat, such as light, moisture, or electric current. Integrating sensing elements or localized heating mechanisms could further facilitate precise, closed-loop control of actuation. Additionally, future research should prioritize optimizing cyclic durability to prevent layer delamination and mechanical degradation over repeated deformation cycles. Implementing these design strategies in large-scale manufacturing could extend application potential to soft robotics, biomedical devices, and smart packaging systems. Ultimately, this study redefines the capabilities of unmodified PLA, demonstrating that intelligent design can transform a static polymer into a dynamic, shape-memory material while upholding PLA's environmental and economic advantages, positioning it as a viable alternative for next-generation smart manufacturing.

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