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# 3D Printing Applications and Development of PLLA: A Review

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## Abstract

**Poly(L-lactic acid) (PLLA), a biodegradable and biocompatible polymer, has emerged as a cornerstone material in additive manufacturing for biomedical applications. This review examines recent advancements in 3D-printed PLLA-based scaffolds and devices, focusing on bone regeneration, infection control, drug delivery, and multifunctional tissue engineering. We analyze innovations in material modifications (e.g., plasticizers, nanocomposites), printing techniques (FDM, SLS, DLP), and structural design strategies (gradient porosity, auxetic architectures). Critical challenges such as degradation control, mechanical optimization, and clinical translation are discussed, along with future directions in 4D printing and personalized medicine.**

## Keywords

**PLLA; 3D-Printing; Additive Manufacturing.**

## 1. Introduction

Polylactic acid (PLA), a linear aliphatic polymer, exhibits semicrystalline characteristics. While it demonstrates favorable impact resistance, this material is limited by its low elongation at break, presenting as a brittle polymer under ambient conditions. Typically synthesized through the ring-opening polymerization of lactic acid (LA) monomers, PLA derives its raw materials predominantly from natural biomass sources such as corn, sugar beet, and straw[1]. In the domain of 3D printing, poly-L-lactic acid (PLLA) has emerged as a material of significant promise. As illustrated in Figure 1 within the polymer history overview, its advantageous attributes-including high structural strength, a controlled degradation rate, and excellent processability-render it highly suitable for fabricating intricate three-dimensional architectures. This review endeavors to provide a comprehensive summary of current PLLA applications in 3D printing, explore state-of-the-art research advancements, analyze existing technical challenges, and project future developmental trajectories.PLLA also showcases notable mechanical performance. With relatively high tensile strength and modulus values, it meets the structural integrity requirements for various engineering applications. For instance, in packaging industries, PLLA can be processed into films and containers that offer adequate strength to safeguard enclosed products. Furthermore, its optical transparency makes it an ideal choice for scenarios requiring visual inspection of packaged items.The processability of PLLA represents another key advantage. It is compatible with conventional polymer processing techniques such as extrusion, injection molding, and blow molding. This compatibility allows industrial adoption without necessitating substantial modifications to existing production lines, facilitating seamless integration into manufacturing workflows.As global demands for sustainable and eco-friendly solutions intensify, PLLA occupies a leading position in material innovation. The unique combination of its biodegradable nature, biocompatibility, mechanical robustness, and process adaptability positions it for expanded utilization across diverse industrial sectors. This article will delve deeper into the synthesis mechanisms, material properties, practical applications, and future prospects of PLLA, highlighting its transformative potential in redefining material usage paradigms.

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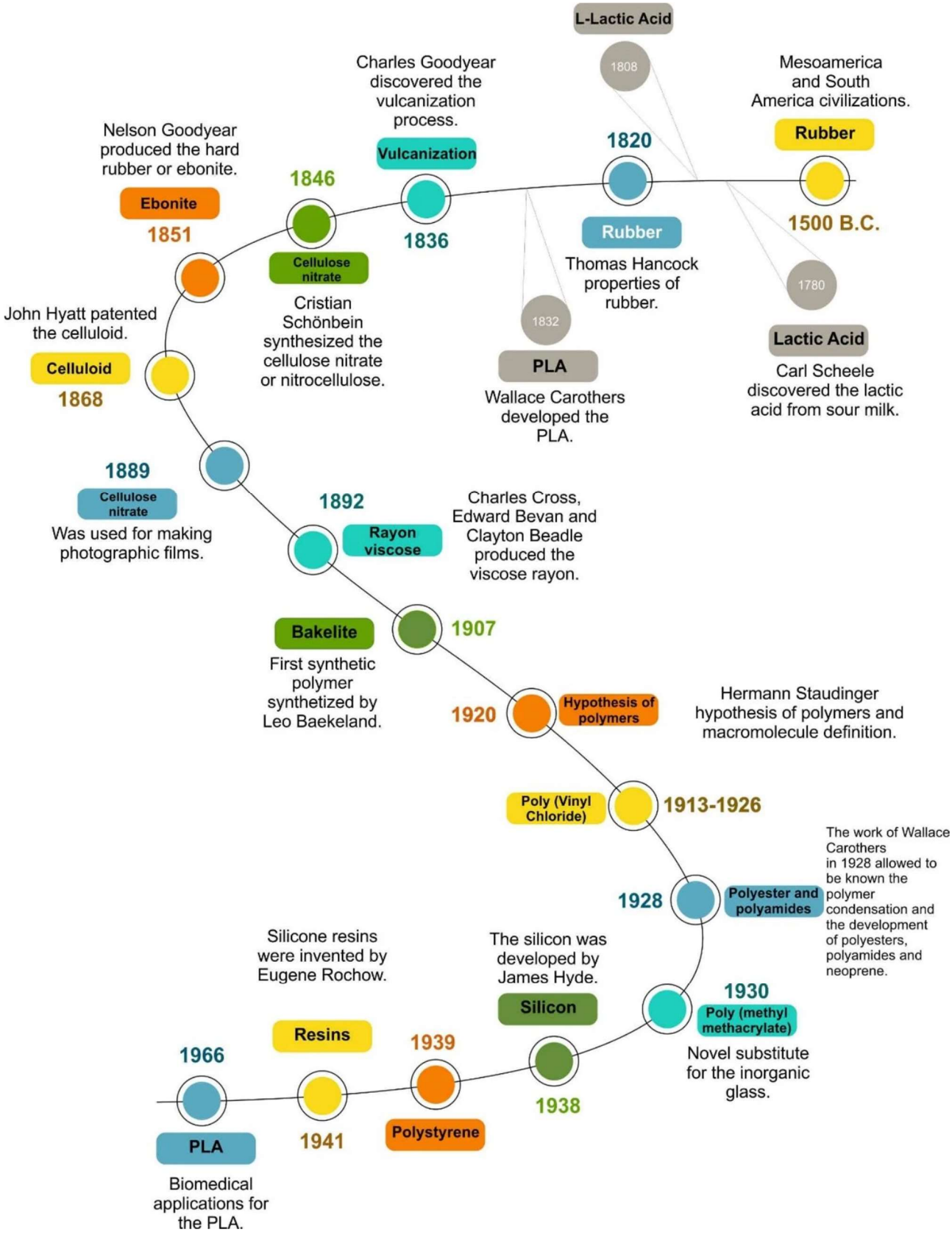


**Figure 1.** Inside the history of polymers[1]

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## 2. 3D Printing Technologies for PLLA

### 2.1 Fused Deposition Modeling (FDM)

Fused deposition modeling (FDM) represents a commonly employed 3D printing method for poly-L-lactic acid (PLLA), where thermoplastic PLLA filaments are heated, melted, and extruded through a nozzle to construct three-dimensional objects in a layer-by-layer fashion. Mincheva et al.[2] utilized FDM to create PLLA-based auxetic materials featuring unconventional re-entrant honeycomb cellular structures. Their approach involved fabricating layered composites with compositional gradients, where successive layers alternated between pure PLLA and plasticized PLLA-with hydrophilic poly(ethylene glycol) (PEG, at 10 and 20 wt% loadings) serving as the plasticizer for the latter. This strategy enhanced the mobility of polyester chains, improved ductility and drawability, and increased hydrophilicity, leading to the development of bone-substituting materials (BSMs) with elevated strength and superior energy absorption capabilities.As showing in figure 2,Cen et al. [3] employed FDM to construct a corn-like PLLA/β-tricalcium phosphate (β-TCP)/chitosan (CS) scaffold, investigating its potential to replace polymethyl methacrylate (PMMA) bone cement in single-stage bone defect reconstruction. The P/T15/S15 scaffolds-containing 15% mass fractions of both β-TCP and CS-successfully induced membrane formation when implanted into large-segmental radius bone defects in white rabbits, demonstrating their feasibility for such applications.Karanth et al. [4] designed and manufactured PLLA scaffolds via FDM, achieving a porosity of 73%, an average pore size of 450 μm, and an average fiber thickness of 130 μm. Notably, these scaffolds generated an electric potential of 25 mV under cyclic/repeated mechanical loading, highlighting their promising utility in bone regeneration applications due to this electroactive behavior.

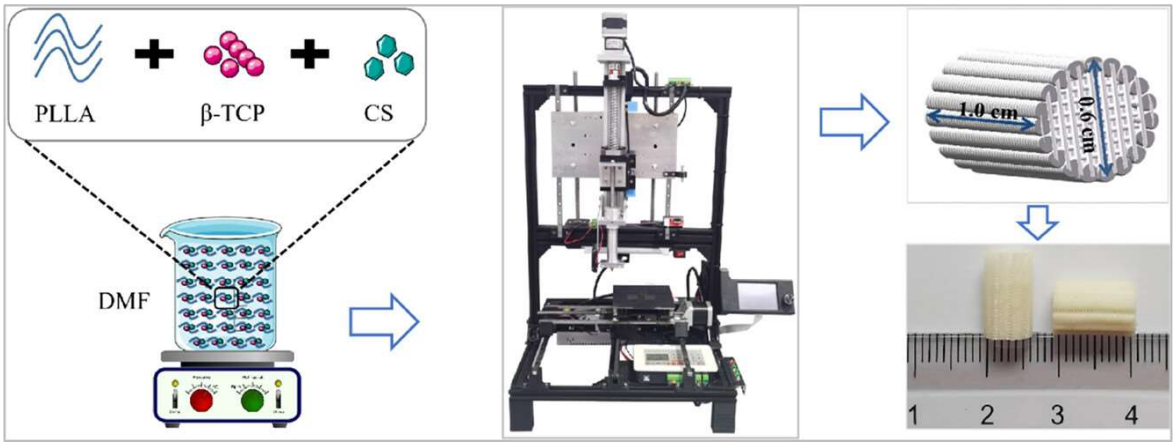


**Figure 2.** Preparation of scaffolds.[3]

### 2.2 Selective Laser Sintering (SLS)

Selective laser sintering (SLS) employs a laser to fuse powdered materials, including poly-L-lactic acid (PLLA), in a layer-by-layer sintering process. Zhao et al. [5] integrated graphene oxide/gallium (GO/Ga) nanocomposites into PLLA matrices and created GO/Ga-PLLA composite scaffolds via SLS. The nanocomposites within these scaffolds exhibit a dual antibacterial mechanism: disrupting bacterial iron metabolism and causing physical membrane damage, making them promising candidates for treating chronic osteomyelitis.We can see in figure 3A,Yuan et al. [6] synthesized carbon–zinc oxide (C–ZnO) nanoparticles and incorporated them into PLLA powder prior to SLS-based fabrication of PLLA/C–ZnO scaffolds. These scaffolds enable sustained release of Zn ions within a biologically appropriate range, enhancing their bioactivity for bone tissue regeneration applications.

### 2.3 Digital Light Processing (DLP)/Stereolithography (SLA)

DLP and SLA are photopolymerization-based additive manufacturing techniques.As showning in figure 3B,Pal et al. [7] developed a biocompatible photocurable resin system comprising acrylate-

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functionalized poly(lactic acid) (PLA) grafted with polyvinyl acetate (PVAc). By blending this polymer with hydroxyl ethyl methacrylate (HEMA) and hydroxyl ethyl acrylate (HEA) as reactive diluents, they produced DLP-printed materials with adjustable mechanical properties through controlled crosslinking.In a related study, Pal et al. [8] designed acrylate-terminated linear and star-shaped poly-l-lactide (PLLA) oligomers as photocurable crosslinkers. Formulated with HEMA as a biodegradable reactive diluent, these systems enabled solvent-free 3D printing using both DLP and a custom direct ink writing setup, showcasing versatility in processing and material design.

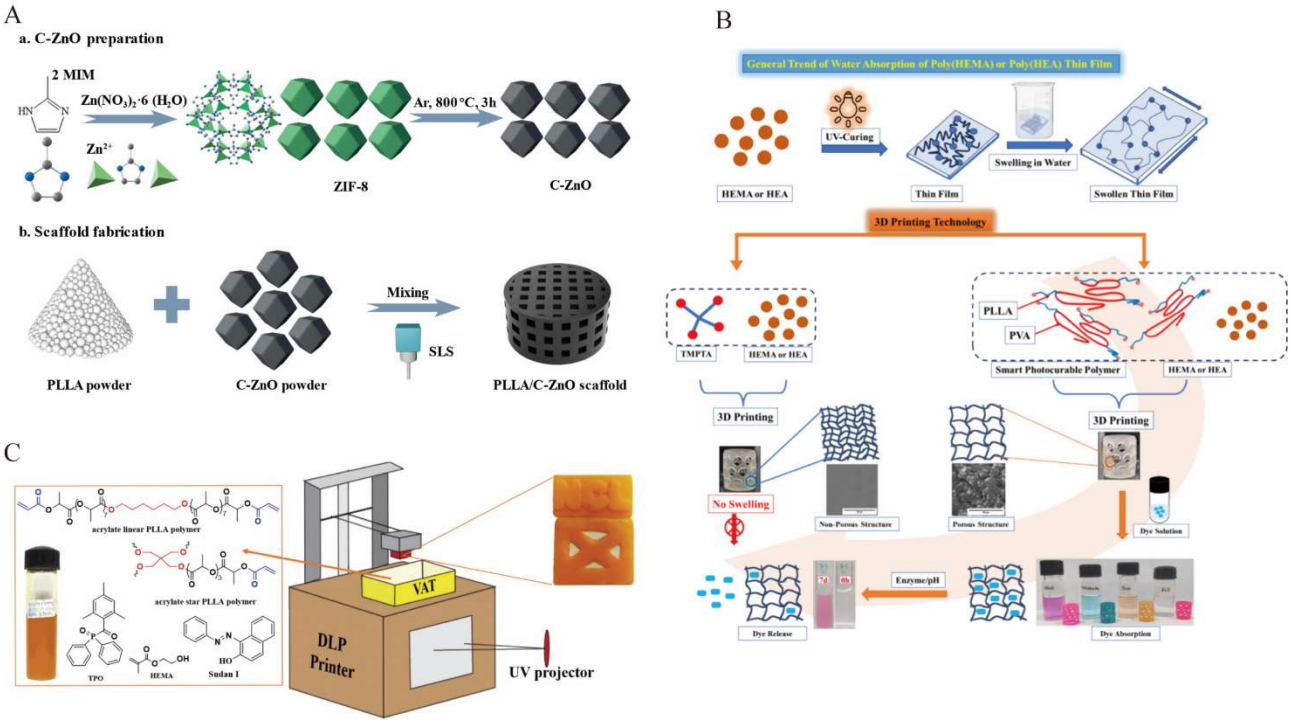


**Figure 3.** A:Schematic illustration of the preparation of C–ZnO scaffolds; B:Schematic representation of the dye absorption and release phenomena of the PVAc-g-PLLA acrylate-based 3D-printed structure. ; C: Schematic representation summarizing the conversion of 3D printable photocurable resin formulation based on PLLA into solid objects using a DLP printer

## 3. Applications for 3D Printed PLLA in Biomedical and Tissue Engineering

### 3.1 Bone Tissue Engineering

3D-printed PLLA scaffolds exhibit significant promise in bone tissue engineering, leveraging precise control over architectural design, porosity, and mechanical properties to facilitate bone regeneration.The auxetic PLLA - based materials developed by Mincheva et al. [1] addressed uniquely mimic natural cancellous bone by addressing previously unreported combinations of porosity, compressive modulus, and impact strength, advancing structural compatibility with native tissues. Cen et al.[3] designed PLLA/β-TCP/CS scaffolds demonstrated efficacy in inducing membrane formation during single-stage bone defect reconstruction, offering a novel strategy for clinical applications.. Khodabandeh et al.[9] engineered microfibrous PCL/HA composite scaffolds via near-field electrospinning (NFES), further incorporating aligned and random PLLA nanofibers. This hybrid architecture enhanced cell adhesion by 334% compared to pure microfibrous scaffolds, with the PLLA nanofibers exhibiting piezoelectric properties that actively promote bone tissue development. De Angelis et al.[10]showed that 3D-printed PLLA+10% hydroxyapatite (HA) scaffolds optimally supported osteoinductivity, facilitating the differentiation of bone marrow-derived mesenchymal stem cells (MSCs) into mature osteocytes. Cho et al. [11] developed BMP-2-conjugated PLLA scaffolds, where controlled release of bone morphogenetic protein-2 enhanced alkaline phosphatase (ALP) activity in MC3T3-E1 and W-20-17 cell lines. This biofunctionalization

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led to a tenfold increase in new bone formation after four weeks compared to unmodified PLLA scaffolds, highlighting the potential of bioactive molecule integration in accelerating osseous regeneration.

### 3.2 Cancer Research

PLLA porous scaffolds offer a valuable platform as three-dimensional tumor models for investigating cancer cell drug resistance. Carbone et al. [12] fabricated PLLA porous scaffolds via thermally induced phase separation and utilized them to construct 3D culture systems for breast cancer cell lines (MDA-MB-231, MCF-7, and MCF-7R). MTS assay results revealed that half-maximal inhibitory concentration (IC50) values were significantly higher in 3D cultures compared to traditional 2D monolayers, indicating that these PLLA scaffolds more accurately mimic the in vivo microenvironment for studying drug resistance mechanisms. Poma et al. [13] evaluated the antimigratory effects of the compound SEMBL within PLLA scaffold-based 3D tumor models. The in vitro findings were validated in these 3D systems, demonstrating that SEMBL exhibits promising antitumor activities in preclinical breast cancer models. This work highlights the utility of PLLA scaffolds as physiologically relevant platforms for assessing therapeutic interventions in a context that better recapitulates tumor complexity.

### 3.3 Wound Healing

Hendrawan et al. [14] loaded conditioned medium from human umbilical cord mesenchymal stem cells (hUC-MSCs) onto 3D collagen-coated PLLA scaffolds (PLLA/CC CM FD) via a freeze-drying technique. When implanted into diabetic rat wounds, these scaffolds promoted enhanced wound closure and increased collagen deposition in the injury site. In vitro experiments revealed that fibroblasts seeded on PLLA/CC CM FD exhibited improved cellular functionality, specifically in collagen synthesis, compared to control groups. This study highlights the potential of bioengineered PLLA-based matrices integrated with stem cell-derived secretomes to enhance tissue repair mechanisms, particularly in challenging diabetic wound environments.

### 3.4 Maxillofacial Surgery

Yamamoto et al. [15] assessed postoperative stability following Le Fort I osteotomy using a u-HA/PLLA composite system, developing a novel multipoint measurement approach for detailed 3D evaluation. The study retrospectively analyzed 31 patients who underwent surgery with Super FIXSORB-MX (composed of u-HA/PLLA), examining maxillary positional changes and postoperative discrepancies through precise three-dimensional measurements.Wang et al. [16] leveraged 3D printing to fabricate customized PLLA porous screws for anterior cruciate ligament (ACL) reconstruction in rabbit knee models. The hydroxyapatite (HA)-modified PLLA-HA screws demonstrated potential to enhance tendon-bone integration, showcasing their utility in promoting biological healing at the interface between the graft and bone tunnel.

### 3.5 Other Biomedical Applications

Zhao et al. [17] introduced a method to fabricate multifunctional personalized sodium alginate scaffolds with improved mechanical stability, enhanced osteogenic potential, and superior anti-inflammatory properties. The 3D printing ink was formulated by dispersing ibuprofen-loaded modified PLLA microdroplets into a sodium alginate aqueous solution. The resulting scaffold exhibited sustained-release behavior for ibuprofen, enabling prolonged anti-inflammatory effects through controlled drug delivery.Du et al. [18] utilized 3D printing to fabricate specimens of the PLLA/IR3535 polymer-repellent system. Experimental results demonstrated the feasibility of printing 3D components containing up to 25 mass% insect repellent, with only minimal repellent loss during the manufacturing process. This system serves as an effective insect-repellent delivery platform, offering potential for combating mosquito-borne tropical diseases through targeted release mechanisms.

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# 4. Modification of PLLA for 3D Printing

## 4.1 Plasticization

Plasticization represents a widely adopted approach to enhance the processability and functional properties of poly-L-lactic acid (PLLA). Mincheva et al. [2] incorporated hydrophilic poly(ethylene glycol) (PEG) as a plasticizing agent into PLLA, demonstrating that the plasticized material exhibited improved mobility of polyester chains, enhanced ductility, superior drawability, and increased hydrophilicity. These property enhancements are particularly conducive to bone regeneration applications, where material flexibility, formability, and biocompatible surface characteristics are critical for cellular interaction and tissue integration.

## 4.2 Incorporation of Nanocomposites

Integrating nanocomposites into poly-L-lactic acid (PLLA) offers a viable strategy to impart additional functionalities to the matrix. Zhao et al. [5] constructed graphene oxide/gallium (GO/Ga) nanocomposites by anchoring gallium nanoparticles onto graphene oxide sheets, which were then incorporated into PLLA. The resulting GO/Ga-PLLA composite scaffolds exhibited robust antibacterial properties, primarily through dual mechanisms: disrupting bacterial iron metabolic pathways and causing physical membrane damage, making them suitable for treating osteomyelitis. Zou et al. [19] designed a polydopamine (PDA)-coated multi-walled carbon nanotube-zinc oxide hybrid nanostructure ((MWCNTs-ZnO)@PDA) and integrated it into PLLA matrices. This nanocomposite system endowed the PLLA with enhanced mechanical strength, improved photothermal conversion efficiency, near-infrared (NIR)-triggered shape memory behavior, and antibacterial functionality. The synergistic effects of MWCNTs-ZnO and PDA coatings not only reinforced the polymer matrix but also enabled multifunctional performance critical for advanced biomedical applications.

## 4.3 Polymer Blending

Polymer blending stands as an effective approach to tailor the properties of poly-L-lactic acid (PLLA) for specific applications. Wu et al. [20] utilized a one-pot synthesis method to create polyvinyl acetate-modified cellulose nanocrystals (CNCs-PVAc) powder, which was incorporated into a PLLA/poly(butylene adipate-co-terephthalate) (PBAT) blend for 3D printing filaments. The inclusion of ungrafted PVAc homopolymer in the composite promoted improved dispersion of CNCs within the PLLA/PBAT matrix, leading to simultaneous enhancements in both mechanical strength and toughness. Hu et al. [21] developed a novel binary substitution material by blending poly(lactic acid-co-trimethylene carbonate) and poly(glycolic acid-co-trimethylene carbonate) with PLLA. This blending strategy not only accelerated the degradation rate of the poly(lactic acid-co-trimethylene carbonate) base material but also enhanced its mechanical properties. The results highlight the potential of copolymer blending to balance biodegradability and structural performance in PLLA-based systems, enabling customized material behavior for diverse applications.

# 5. Degradation of 3D Printed PLLA

Malone et al. [22]conducted a study on the degradation mechanisms of 3D-printed amorphous poly-L-lactic acid (PLLA) fibers, subjecting samples to fluid environments at 37°C, 50°C, and 80°C over a six-month period. Results indicated that across all tested temperatures, the fibers underwent bulk homogeneous degradation. At elevated temperatures, PLLA crystallization occurred, and a three-stage degradation process was identified through analysis of fluid pH changes, fiber mass loss, molecular weight fluctuations, and polydispersity index variations. These measurements provided insights into the sequential chemical and physical transformations during degradation.Duan et al. [23]evaluated the in vivo biodegradation and biocompatibility of 3D-printed scaffolds composed of PLCL/PLLA blends. While all scaffolds exhibited acceptable biocompatibility, the PLCL50/PLLA50 scaffold—containing the highest PLLA proportion—induced the thickest fibrous capsule formation around the implant and displayed the highest inflammatory response scores. This suggests that PLLA

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ratio significantly influences the host tissue reaction, highlighting the need for optimized blend compositions to balance biodegradability and biocompatibility in clinical applications.

## 6. Conclusion

The 3D printing of poly-L-lactic acid (PLLA) has emerged as a transformative technology with wide-ranging applications across biomedical and tissue engineering domains. Leveraging diverse additive manufacturing techniques, PLLA can be fabricated into intricate architectures with customized properties, catering to specific clinical and engineering requirements. Modification strategies-such as plasticization, nanocomposite incorporation, and polymer blending-further augment its functional versatility, enabling tailored performance in applications from bone tissue engineering to drug delivery systems. Notwithstanding these advancements, several challenges persist. Future research should prioritize the development of novel material formulations, optimization of printing processes to enhance structural precision, and addressing unresolved issues related to degradation kinetics and long-term biocompatibility. By fostering continuous innovation in material science and manufacturing technology, 3D-printed PLLA is poised to assume an increasingly pivotal role in advancing next-generation medical devices and regenerative therapies, driving breakthroughs in personalized medicine and sustainable bioengineering solutions.

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