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Shape Memory Polymers in Tissue Engineering Scaffolds

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Abstract. In recent years, driven by the pursuit of minimizing surgical trauma, minimally invasive procedures and related medical techniques have advanced rapidly. Among the medical devices employed in such procedures, shape memory polymer (SMP) scaffolds have gained widespread application in tissue repair due to their shape memory effect, high biocompatibility, and degradability. This study analyzes and compares the performance and fabrication processes of different SMP scaffolds, along with the impact of functionalization modifications on degradation properties. Several typical application cases are examined. Finally, the limitations of SMP scaffolds in tissue repair and potential improvement directions are discussed. This study provides a systematic theoretical basis and technical reference for the development of a new generation of high-performance tissue engineering scaffolds. The results have important guiding value for promoting the personalized design and clinical application of minimally invasive repair materials for translation. In the future, the results can be used to break through the key technological bottlenecks in the precise control of degradation rate and optimization of biological activity.

Keywords: Shape-memory polymer (SMP); scaffold; 4D printing; tissue repair.

1. Introduction

With continuous advancements in medical technology, people have set higher expectations for modern surgical procedures, particularly in minimizing the physical damage caused by operations. Among these, “minimally invasive” techniques represent a crucial aspect. SMP materials exhibit superior biocompatibility, lightweight properties, biodegradability, and shape memory compared to traditional materials, making them a research hotspot for developing implantable tissue repair scaffolds. Although implantable scaffolds based on SMP materials are currently developing rapidly, there remains room for optimization in aspects such as scaffold preparation, functionalization modification, and biodegradability. For example, the methods used to prepare stents from different SMP materials exhibit significant variations, which greatly impact the performance of the finished stents. Additionally, modifying the stents can also affect their original properties. To advance the development of SMP materials for fabricating implantable tissue repair scaffolds, this study will primarily review the following aspects:First, differences in SMP scaffold preparation processes and their resulting properties; second, the impact of functional modifications on degradation performance; and finally, illustrative examples of SMP scaffolds' applications in various tissue repair scenarios. This study aims to summarize and compare the existing fabrication processes of SMP scaffolds, the effects of functionalization modifications, and specific case studies of their application in different tissue repair scenarios, thereby providing a reference for subsequent research.

2. Basic Concepts and Current Development Status of SMP Materials

2.1. Overview of SMP Materials

SMP materials refer to polymer materials with shape memory functionality. During processing, applying a certain degree of pre-deformation to the SMP material enables it to partially revert to its pre-deformed state when subjected to a specific stimulus after the material has been shaped. Broadly speaking, polymers with similar functions can be considered SMP materials. The shape memory effect refers to the ability of polymers to undergo reversible transitions between a temporary shape

and a permanent shape when stimulated by external fields (which may be single or multiple stimuli, such as sound, light, heat, force, or electricity). The essence of this capability stems from the dual-component nature and entropy-driven behavior of the polymer network structure [1]. Additionally, it is worth noting that Huang et al. believe that the shape memory behavior of polymers is closely related to their glass transition behavior. Therefore, they point out that the vast majority of polymers (except for those with highly oriented segments like Kevlar fibers) exhibit some degree of shape memory capability [2].

2.2. Preparation Methods and Typical Characteristics of Support Structures for Common SMP Materials

2.2.1. Polyurethane (PU)

In-situ polymerization/prepolymerization method, wherein soft segments and hard segments polymerize under the action of crosslinking agents to form PU; it can be further composite with cellulose nanofibers (CNFs), hydroxyapatite (HA), etc. The scaffold materials prepared by this method allow for free ratio adjustment, controllable chemical structure, and flexible molecular segment design [3][4].

The electrospinning method can mix PU solutions with various nanoparticles to electrospin them into nanofiber membrane scaffolds. The electrospinning process for preparing scaffolds is simple, allowing fiber diameter control through voltage adjustment and yielding scaffolds with high porosity [5].

3D printing utilizes TPU or biodegradable PU as printing materials, enabling programmable porous structures through CAD and other design software. Some studies incorporate composite phases. The most prominent advantages of 3D printing are structural customizability and excellent interconnected porosity. Depending on the composite phase added, scaffolds suitable for different tissues can be designed [6-8].

Solvent casting/foaming & freeze-drying method: Typically, PU scaffolds with porous structures are prepared through solvent foaming or salt immersion combined with freeze-drying. The pore structure of scaffolds produced by this method exhibits significant randomness. Although pore size distribution can be controlled through process adjustments [8], precise regulation like that achievable with 3D printing is not possible, resulting in considerable limitations. Table 1. Comprehensive comparison of the performance and structural differences among PU scaffolds prepared using various methods.

Table 1. Performance Comparison of PU Stents Prepared by Different Processes

Preparation Method	Structural Features	Shape Retention Rate % (Rf)	Shape Recovery Rate % (Rr)	Typical Performance
Prepolymerization Method (PCL-IPDI-GL)	Cross-linked network, body temperature-activated	~95	~96	Recoverable strain up to 450%
CNFs-PCL Grafted + PU CNFs-PCL Grafted + PU	Composite Network	~92	~90	Strength enhanced, toughness improved
3D Printing (TPU/PVDF)	Pores 300–500 μm, piezoelectric	98.6	81.2	Piezoelectric coefficient close to bone tissue

3D Printing (Selective Laser Sintering)	Water-responsive pore	67	90	Body temperature triggers cell-directed proliferation upon immersion in water.
Foaming + Freeze-Drying	Programmable Pores, HA-Enhanced	>98.9	>96.2	Excellent biodegradability with significant bone regeneration
3D Printing + Mg Composite	Close contact, NIR response	93.6	95.4	Light-and-heat-activated, enhancing load capacity
Water-based synthesis + 3D printing	Containing SPIO nanoparticles	~90	~92	Magnetic Response & Osteogenic Induction
Electrospinning + HA	Nanofiber network	~94	~91	Mechanical properties enhanced, high porosity

2.2.2. Polylactic Acid (PLA)

Table 2. Performance Comparison of PLA Scaffolds Prepared Using Different 3D Printing Methods[9-12]

Preparation Method	Structural Features	Performance Metrics	Biocompatibility
Fused Filament Fabrication (FFF) 3D printing, incorporating Wollastonite ceramic particles into a PLA/PCL matrix	Triple-phase composite scaffold with controllable porosity and significantly enhanced compressive strength (approximately twice that of pure PLA).	Shape recovery ratio ≈ 84%	Cells exhibit good adhesion and proliferation, and possess bone-inducing activity.
3D Printing Combined with Mg Particle Doping to Construct Photothermal Composite Scaffolds	The porous structure incorporates Mg particles that impart near-infrared (NIR) light responsiveness; the scaffold exhibits rapid shape recovery at 50°C.	Shape retention and recovery rates are both close to 100%; Response speed is fast (restores within seconds).	Cells exhibit robust viability, promote osteogenic differentiation, and demonstrate certain antitumor potential.
Selective Laser Sintering (SLS), PLA-TPU blend, incorporating ZnO@MWCNT conductive assemblies	Porous conductive network capable of shape memory driven by electrical stimulation	Fixed rate: 98.0%, Recovery rate: 98.8% (under electrical stimulation conditions)	High biocompatibility, supporting osteoblast adhesion and proliferation
3D Printing + Surface Modification (Plasma Treatment + Collagen Coating + Hydroxyapatite Mineralization)	Multi-layered biomimetic structure with enhanced surface roughness for improved bone repair functionality	The PLA matrix exhibits a certain degree of thermally triggered reversibility.	Significantly enhanced biocompatibility promotes osteoblast adhesion and differentiation.

As shown in Table 2, the current mainstream methods for preparing PLA scaffolds are 3D printing and composite modification. However, multi-stimulus responses to light/electricity/magnetism, such as novel preparation methods like Mg particles enabling photothermal triggering and ZnO@MWCNT constructing electrically driven networks, show great promise. Regarding scaffold performance, the PLA/PCL-Mg, NIR-triggered, and PLLA/TPU conductive SLS scaffolds exhibited the highest shape retention/recovery rates, approaching 100%.In terms of response speed, the NIR light-triggered system can achieve response times measured in seconds. In terms of mechanical properties, PLA/PCL/Wollastonite exhibits approximately twice the compressive strength of pure PLA. In terms of biocompatibility, modifying the scaffold significantly enhances osteocyte adhesion and osteogenic induction capacity.

2.2.3. Polycaprolactone (PCL)

Currently, a combination of fused deposition modeling and electrospinning techniques is primarily employed to fabricate multiscale PCL scaffolds. The standard preparation process is as follows: First, a PCL scaffold is printed using FDM technology to form the base structure. Electrospinning is then performed on top of the base to create a nanoscale fiber layer. This process is repeated to form a multilayer structure. For example, a PCL scaffold prepared using this method has a diameter of 21 mm and a height of 1 mm. It features a multilayer structure comprising micrometer-scale printed layers and nanometer-scale electrospun layers. The electrospun layer fibers have a diameter of approximately 500 nm. The scaffold exhibits a porosity of about 70%. Mechanical property testing indicates that the scaffold exhibits excellent compressive strength and elastic modulus. Cell culture experiments demonstrate that the scaffold supports cell adhesion and proliferation [13].

Thermoresponsive SMP scaffolds were prepared using UV-crosslinkable PCL macromonomers (Star-PCL). Star-PCL was mixed with a crosslinking agent to form a photosensitive solution. The solution was cast into a mold and crosslinked via UV irradiation, yielding scaffolds with shape memory properties. This scaffold features an adjustable transition temperature. By modifying the molecular weight and crosslinking degree of Star-PCL, the transition temperature range can be regulated. This range varies from 37°C to 55°C. The scaffold can revert to its preset shape at specific temperatures. It exhibits excellent mechanical properties, making it suitable for bone tissue engineering [14].

3. The Effect of Functionalization on Degradation Behavior of SMP Materials

3.1. PLA/PCL as the matrix

The degradation behavior of different SMP material systems varies after modification, reflecting the regulatory role of material chemical composition and structural design in degradation mechanisms. Taking the PLA/PCL system as an example, Arabiyat et al. significantly accelerated the degradation rate of scaffolds in PBS by introducing PLLA into the PCL-DA network to form a semi-interpenetrating polymer network (semi-IPN), supplemented by poly(dopamine) surface functionalization [15]. As shown in Figure 1.a), compared to the pure PCL-DA scaffold, which exhibits significant degradation only after more than two years, the PCL-DA/PLLA semi-IPN demonstrates approximately 3% mass loss within 14 days. The primary reason lies in the limited compatibility between PCL and PLLA, which leads to phase-separated structures being more prone to water absorption, thereby accelerating the hydrolysis process. These results indicate that utilizing blending or semi-interpenetrating networks can effectively shorten the degradation cycle of polyester-based SMPs, offering the potential to match tissue repair rates.

3.2. PU as the Matrix

In PU-based SMPs, the role of functionalization manifests as programmable regulation of degradation mechanisms. Vakil et al. reported a series of polyurethane SMP foams prepared by modifying the chemical composition of the main chain and introducing oxidizable groups [16]. As shown in Figure

1.c), under accelerated oxidation conditions (3% H_2O_2, 37°C), the material completely degraded within 30–60 days. The degradation curves exhibited significant differences among different components, indicating that chemical functionalization enables tunable degradation rates. Further studies indicate that foams enhanced with hydrophilicity and incorporating dual-mechanism groups, as shown in Figure 1.b), exhibit in vitro degradation curves under 1% H_2O_2 conditions that closely match the 12-week in vivo degradation observed in rat subcutaneous tissue. In contrast, hydrophobic formulations dominated by oxidative degradation require 3% H_2O_2 in vitro conditions to simulate their in vivo degradation [17]. This functional regulation not only endows PU-based SMPs with precisely controllable degradation behavior but also provides a basis for establishing in vitro-in vivo correlations.

Comparing the two examples above reveals that the functionalization of the PLA/PCL system relies more heavily on compatibility regulation and surface modification to accelerate degradation, whereas the PU system achieves precise rate control across a broader range through molecular structural design. This indicates that functionalization strategies for different SMP scaffolds should be tailored to their intrinsic chemical properties: polyester-based materials are better suited for accelerated degradation through blending/interface engineering, while polyurethanes can achieve tunable degradation via functional group design. Overall, functionalization modifications provide multiple pathways to achieve the design principle of "matching scaffold degradation rates with tissue regeneration rates," laying the groundwork for subsequent clinical translation.

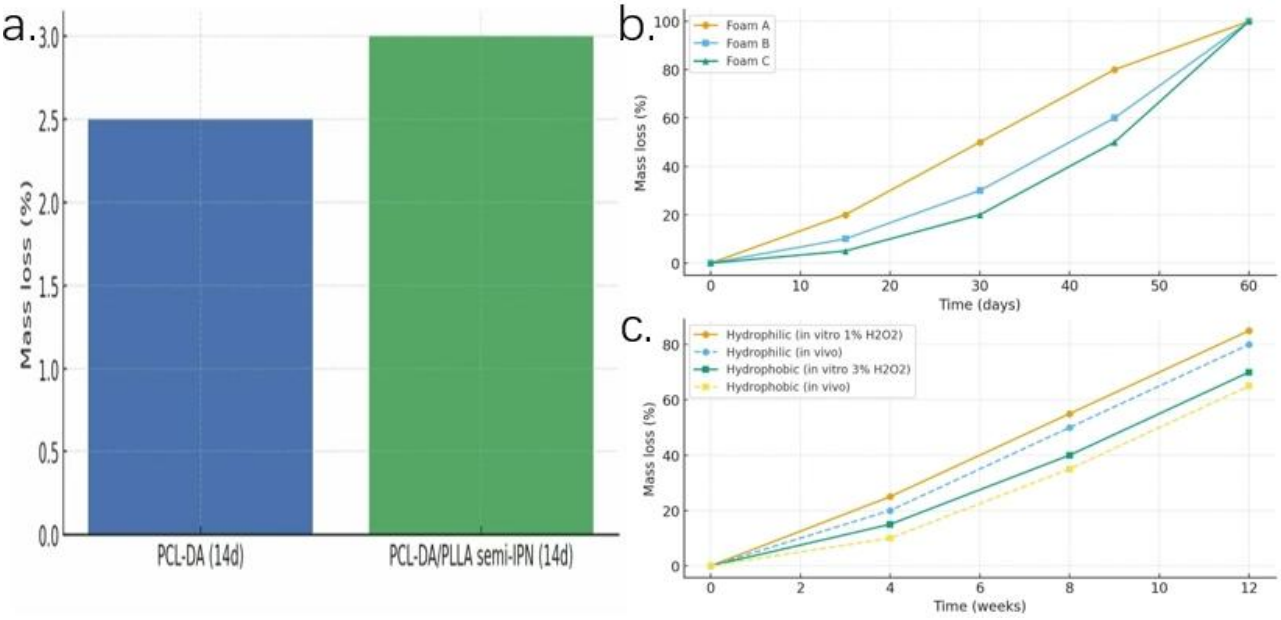

Figure .1 a) Comparison of mass loss after 14 days of PBS degradation; b) Three formulations under accelerated degradation conditions (complete degradation within 30–60 days); c) Comparison of highly hydrophilic/hydrophobic formulations in vitro H_2O_2 and in vivo rat experiments.

4. Analysis of 4D-Printed SMP-Based Smart Scaffolds for Various Tissue Types

4.1. Bone Tissue

Zhao et al. constructed a multi-responsive bilayer membrane structure using 4D printing technology, comprising a PCLDA layer and a hydrogel layer [18]. The PCLDA layer features temperature-responsive surface microstructures that transition from a smooth state to a roughened structure upon body temperature stimulation, thereby regulating the transition of stem cells from proliferation to osteogenic differentiation. The hydrogel layer achieves macroscopic shape adaptation through swelling, conforming to complex bone defect contours. In vivo experiments demonstrate that this scaffold significantly promotes new bone formation in a mouse femoral defect model. Compared to

static scaffolds, it increases bone volume fraction (BV/TV) by over 30% and enhances the expression of osteogenesis-related genes (RUNX2, OSX) by activating calcium ion channels through remote stimulation.

4.2. Soft Tissue (Fat)

Andrew P. Dove et al. developed a 4D-printed scaffold based on aliphatic polycarbonate, fabricated using stereolithography [19]. This material exhibits outstanding shape memory properties, with a shape recovery rate exceeding 95%. It also possesses unique surface erosion characteristics, maintaining mechanical stability throughout its in vivo degradation process. After implantation into mouse models, the scaffold restored its pre-set porous structure upon body temperature activation, promoting adipocyte infiltration and angiogenesis. Within two months, adipose lobules and neovascularization invaded the scaffold interior, accompanied by reduced collagen capsule thickness, indicating minimal inflammatory response.

4.3. Neural Canal

4D-printed SMP neural conduits can dynamically deform to match the neural regeneration process. For example, PCL-based neural conduits can gradually expand in vivo alongside nerve axon growth, providing directional mechanical guidance. They activate voltage-gated calcium channels through electrical stimulation, thereby promoting Schwann cell migration and myelination. Experiments have demonstrated that the 4D-printed group enhances nerve regeneration efficiency by approximately 12% compared to static catheters (animal model data) [20].

4.4. Cardiovascular-Related Tissues

4D-printed bio-piezoelectric scaffolds combine the deformation capability of SMP with the electrical activity of piezoelectric materials. For example, PLA/PCL composite scaffolds generate surface potential changes (~100 mV) under ultrasonic stimulation, mimicking the myocardial electrical microenvironment and promoting synchronized contraction of cardiomyocytes. In rat experiments, the myocardial infarction area in the group implanted with 4D piezoelectric patches showed a 20% reduction in cell apoptosis rate and a significant increase in vascular density [21].

5. Conclusion

This study summarizes the preparation processes and performance comparisons of three types of SMP scaffolds—PU, PLA, and PCL—analyzes the regulatory effects of functionalized modifications on their degradation behavior, and explores their applications in various tissue repair scenarios. Overall, SMP scaffolds demonstrate unique advantages in shape memory performance, biocompatibility, and degradability. However, existing research still faces challenges such as unclear mechanisms, imperfect evaluation systems, and insufficient clinical translation validation. Meanwhile, this study also has limitations in terms of functionalization and application coverage. Future research should focus on investigating the coupling mechanisms between scaffold degradation and tissue regeneration, promoting the application of new technologies such as multi-material composites and 4D printing, and strengthening systematic animal experiments and preclinical validation. This will lay the foundation for the widespread application of SMP scaffolds in regenerative medicine.

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