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Review

Shape Memory Polymeric Materials for Biomedical Applications: An Update

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Abstract: Shape memory polymers (SMPs) are emerging smart materials that have the ability to change to a shape and revert to their permanent shape on application of external stimulus. SMPs can be divided into four types based on their permanent shape, elasticity origin, and temporary shape fixing mechanism: chemically cross-linked glassy thermosets, chemically cross-linked semi-crystalline, physically cross-linked glassy copolymers, and physically cross-linked block copolymers. The broad overview of recent developments on SMPs for biomedical applications proves their wide applicability in the field of general medicine, drug delivery, regenerative medicine, dentistry, neuromedicine, cancer therapy, orthopedics, and corrosion protection. Herein, a comprehensive analysis of SMPs and their composites with focus on their types, mechanism, functionality and biomedical applications is presented. Relevant biomedical technologies based on SMPs and their future advancement towards biomedical applications are also discussed.

Keywords: shape memory materials; smart polymers; shape memory polymer; composites; medicine; dentistry

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1. Introduction

Polymeric materials have replaced traditional materials for various products due to their light weight, low cost, high strength, ease of processability and resistance to chemicals and harsh environments [1–3]. Continuous attempts have been made toward the development of new polymeric materials that could serve a wide range of present-day applications [4–6]. The growing advantages of versatility, design freedom, and tunable properties, in addition to their readiness for scaling up from lab to the industrial scale, favors the development of polymeric materials [3]. Meanwhile, the needs for the improvement in polymeric materials performance for different applications has led to the development of engineered materials including polymeric blends and micro/nano composites [7,8].

Shape memory polymers (SMPs) are the emerging smart materials that can change shape temporarily and revert to their shape upon external stimulus or command. SMPs are increasingly investigated due to their favorable properties such as light weight and low density, good flexibility and ease of processability. In addition, SMPs possess tailorable physical properties such as porosity, orientation, and microstructrures that can be adjusted to mimick biological materials such as native extracellular matrix [9–11]. For instance, SMPs in micro/nanofiber form have been widely used in the biomedical field as they show dynamic fibrous structures that are suitable to guide cell regulation and differentiation, control drug delivery, and/or guarantee scaffold [12,13]. Another interesting application area for varieties of SMPs and their composites is due to the possibility to alter their physical and chemical compositions to tailor intrinsic thermal transitions [14,15] within the range of needs of biomedical applications. This review focuses on the development and advanced state-of-the-art in SMPs and their composites, with focus on their types, mechanism, functionality and biomedical applications. Relevant biomedical technologies based on SMPs and their future advancement towards biomedical applications are also discussed.

1.1. Shape Memory Polymers (SMPs)

SMPs are emerging smart materials that can change their shape temporarily and revert to their shape upon external stimulus or command. The shape memory effect (SME) of polymers can be achieved by the combination of their compositions, physical properties such as thermal transitions, and processing or programming conditions [16]. For instance, other approaches that involve merely programming by deforming at ambient temperature and then letting it be relaxed under the application of stress demonstrate that SME can also be generated without the application of heating/cooling cycles [17].

1.1.1. Structures and Mechanism of SMPs

The general structure of the SMPs is the mixture of appropriate molecular network architecture as shown in Figure 1. The molecular network architecture consists of net points connected by chain segments and switches that respond to external stimuli [18]. When combined with a suitable/tailored programming stimulus, the chain segments undergo elastic deformation that can be reverted back to the original structure or shape by reorienting the chain segments [19,20]. The transition temperature (T_{trans}), above which the elastic deformation of the network occurs, facilitates the chain segment deformation and can be fixed in a temporary or deformed shape. Netpoints that dictate the shape of the polymeric network can undergo physical (e.g., intermolecular interactions or phase separation) or chemical (e.g., reversible covalent bonds) changes. Figure 1A shows an example of a SMP with thermal programming. Heating the polymer above the $T_{trans,}$ enables its deformation, acting as a molecular switch by melting the glassy/crystalline phase, thus creating a temporary shape that can be fixed by cooling the material below the T_{trans}. The original shape is achieved again by heating the materials above the same T_{trans}.

The network architectures essential for material to show SME are shown in Figure 1B [21]. The molecular switches and netpoints are arranged in four structures: switching segments linking netpoints, side chains as switching segments, molecular switches forming covalent bonds reversibly, and triblock segments (ABA) linking netpoints. Cross-linking of polymer network covalently, aided by switching domains that acts as the linking netpoints of the network, thereby creating one or two phases depending on the glass transition (T_g) and T_{trans} [19,22]. The polymeric network architecture can also have additional side chains, which can be miscible or segregate into different phases and thus creating multiphase switching segments. Functional groups (e.g., photo-reversible) can also facilitate molecular switches as they can form and reversibly split covalent bonds when exposed to appropriate external stimuli [18]. Multiphase morphology can also be formed by different segments, as in the case of triblock structures, and can act as switching domains with one or multiple blocks [23].

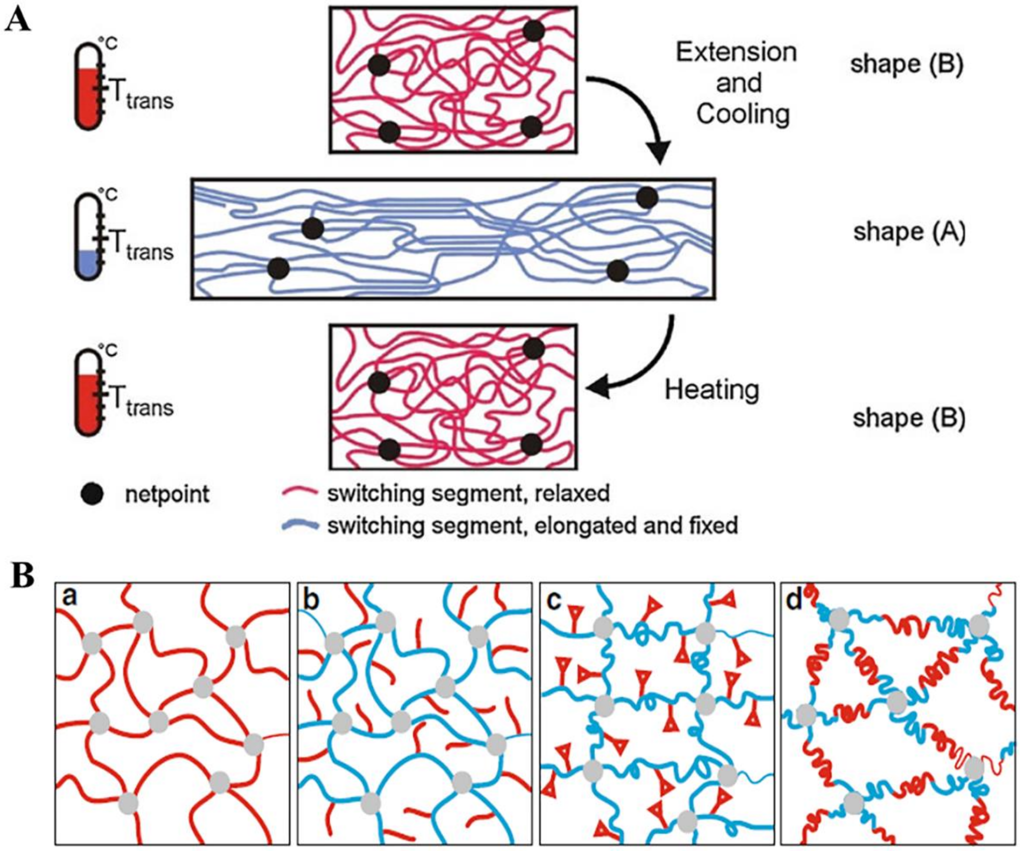

Figure 1. Shape memory effect of polymeric materials. (**A**) Thermal programming of shape memory effect (SME) [24] and (**B**) Polymer network showing SME (red = molecular switches and gray = netpoints): switching segments linking netpoints (**a**), side chains as switching segments (**b**), molecular switches forming covalent bond reversibly (**c**), ABA triblock segments linking netpoints (**d**) [21]. Reprinted with permission from [21,24]. T_{trans} = thermal transition temperature of the switching.

The SMPs from the different classes mentioned before are generally actuated by using heat as an external stimulus. However, apart from heat, the design of SMPs have expanded the stimulus methods to light, moisture, pH, and magnetic and electric fields [25].

Thermal stimulus: The thermal stimulus employs the intrinsic temperature transitions such as glass transition and melting or liquid crystalline transition of the polymer networks to enable shape memory behavior. Block copolymers such as PU, polyetheresters have their $T_{trans} = T_m$ [26,27], and T_m is used to create and fix the temporary shape. Amorphous polymer networks or mono-component polymer networks have $T_{trans} = T_g$ [28] and for such materials, T_g is used for SME. The thermal transition of the liquid crystalline moieties in liquid crystalline elastomer (LCE) networks are also used to enable thermally-induced SME [29]. Slow shape recovery and materials that have their T_{trans} near to the physiological body temperature are attractive for biomedical applications.

The Indirect thermal stimulus for the SME can be achieved by indirect heating, e.g., by the electric field, irradiation, or magnetic field. Thermally-stimulated SMPs can be heated by irradiating with infrared light e.g., laser-activated pUs [30,31]. The addition of magnetic nanoparticles such as Fe(III)oxide cores and Ni-Zn ferrite particles and other functional fillers in thermoplastics affords remote thermal actuation of the SME using alternating magnetic fields [32,33].

Light as a stimulus: SME can be induced by the incorporation of light-responsive groups with reversible molecular switches such as cinnamic acid or cinnamyliden acetic acid in the polymer network [18,34]. Different wavelength lights can be used for the fixation of permanent shape recovery.

Water/pH as a stimulus: The use of moisture, water and pH as stimuli to trigger shape memory in polymeric materials eliminates any temperature-induced unfavorable effects and is close to human physiological conditions and thus are more advantageous for biomedical applications. Recently it was reported that moisture can be used for water-triggered SMP. Other examples include chemically cross-linked poly(vinyl alcohol) (PVA) and moisture-sensitive polyurethane.

1.1.2. Classification of SMPs

SMPs can be divided into four categories based on their permanent shape elasticity origin and temporary shape fixing mechanism [35]. They are categorized into four groups as follows.

Chemically cross-linked glassy thermoset networks: These are the simplest SMPs and are simply cross-linked polymers that feature a sharp T_g and excellent rubbery elasticity above this T_g. The shape memory characteristics can be precisely altered by varying the degree of cross-linking but after re-shaping, the permanent shape is covalently fixed. Owing to their rubber elasticity, this class of materials displays attractive shape memory characteristics such as an excellent degree of shape recovery. The nature of permanent cross-linking affords the rubber elasticity in such shape memory network and the rubbery modulus in these networks can be adjusted through the extent of covalent cross-linking, thereby allowing tunable recovery capacity [36]. Nevertheless, after being processed once, these materials are difficult to reshape, as the permanent shape is covalently fixed. The shape memory characteristics can be precisely adjusted by varying the extent of cross-linking [35,36]. As these polymers have high molecular weight, they have higher chain entanglements, and these entanglements serve as a physical cross-link forming a 3D polymer network. This gives the polymer excellent rubber elasticity above the T_g and thus the shape recovery. The temporary shape can be very well fixed by vitrification when cooled below the glass transition. Examples include polynorbornene, poly(methyl methacrylate), methyl methacrylate, and the butyl methacrylate monomers with tetraethylene glycol dimethacrylate, etc. [3,26]. However, the high molecular weight is one of the disadvantages of such polymers, as it limits the processing of such materials. Other disadvantages of these materials are that the transition temperature cannot be easily varied and the polymer creeps under stress due to the finite lifetime of the chain entanglements at high temperature.

Chemically cross-linked semi-crystalline networks: These polymeric materials are fixed to temporary shapes by crystallization, instead of vitrification, and the permanent shapes are the result of chemical cross-linking. The melting transition of semi-crystalline components can also be used to trigger shape memory behavior. This class of materials has sharp transition zones and their stiffness is sensitive to the degree of crystallinity, which in turn is dependent on the extent of cross-linking [37]. Bulk polymers, such as semi-crystalline rubbers and liquid-crystal elastomers, are included in this class of materials. Apart from bulk polymers, hydrogels with phase-separated crystalline microdomains also belong to this class of materials [38]. Semi-crystalline rubbers are more favored as shape memory materials (SMMs) due to the fast shape recovery, owing to their superelastic nature and flexible modulus at the fixed stage [39]. Examples include semi-crystalline rubbers and liquid-crystal elastomers (LCEs), peroxide cross-linked trans-polyisoprene (TIP), hydrogels with phase-separated crystalline microdomains, etc. [40,41]. Crystallization kinetics and thermal conductivity affect the shape fixing and recovery properties in such materials. Besides the above-mentioned factors, cross-linking is one more factor, affecting the crystal formation, which might lead to less crystallinity, broader crystal-size distribution, and melting-temperature zone in a semi-crystalline material [42,43]. Recently a successful effort to specifically cross-link only the amorphous fraction was reported which enabled the maintenance of a sharp melting transition and avoided any side effects due to cross-linking [15].

Physically cross-linked glassy copolymers: These polymers exhibit rheological properties like those of conventional thermoplastics required for facile processing. In addition, these polymeric materials usually are phase-separated block copolymers, wherein the amorphous or crystalline domains have super-T_g elasticity [44]. As these materials are only physically cross-linked, they can be re-processed. Examples include semi-crystalline homopolymers, low crystallinity polymers, melt-miscible and compatible polymer blends with semi-crystalline constituents such as polylactic acid, poly(ketone-co-alcohol), Poly(1-hexadecene)-co-PP, etc. [45,46]. These SMPs also include some low crystallinity, semi-crystalline homopolymers, or melt-miscible and compatible polymer blends with at least one semi-crystalline component [47]. In such materials, the crystals serve as hard domains, and T_g of the amorphous fraction functions as the transition temperature. For the miscible polymer blends, the T_g temperature of the amorphous phase and thus the shape recovery can be easily tuned by changing the blend composition. Apart from crystalline and glassy domains, other types of physical cross-linking such as hydrogen bonding or ionic clusters within the hard domains can also help to set the network [48]. These interactions assist in strengthening the hard domains and decreasing the chain slippage during deformation, and hence increasing the extent of shape recovery.

Physically cross-linked semi-crystalline block copolymers: These polymeric materials also include some block copolymers that are semi-crystalline in nature and have segmented morphology consisting of soft segments (flexible, melt-crystallizable) and hard segments (amorphous). The shape is determined by the crystallization of the soft domains when cooled below the melting temperature (Tm) of the soft segments. Examples includes styrene–trans-butadiene–styrene (STBS), polystyrene (PS), poly(trans-butadiene) (TPB), polyurethanes (PU), etc. [49,50]. The hard segments form physical cross-links by way of polar interaction, and hydrogen bonding, and such cross-links capable of withstanding moderately high temperatures. The soft segments are melt-crystallizable forming the thermally reversible phase and the melting/crystallization of these soft segments creates/fixes the temporary shape. The properties of polyurethanes such as stiffness, transition temperature, and shape memory, can be easily tuned by manipulating their compositions.

Along with the above-mentioned shape-memory materials, other existing types of materials displaying shape memory characteristics are the gels and liquid crystalline elastomers described below.

Polymer Hydrogels: Shape-memory gels are also cross-linked materials analogous to shape-memory rubbers. A typical shape-memory gel consists of a hydrophilic fraction and hydrophobic sections. The hydrophilic section can be swelled in water, while the hydrophobic section can undergo temperature-controlled, reversible order-disorder of the structures.

Reversible shape memory elastomers: These SMMs are widely used for the applications such as reversible actuators, especially for artificial muscles, which are growing. Such applications require materials that can withstand highly reversible strains up to 30%, a stress of 350 kPa, and fast actuation. A wide range of materials such as the modulated gels discussed before, electroactive-conducting polymers [51,52], and liquid-crystalline elastomers (LCEs) [53,54].

SMPs can be also classified according to their SME, as one-way SME (1W-SME), two-way reversible SME (2W-SME) tripe SME (triple-SME), and multiple SME as below:

One-way SME: An external force is required to change the shape and the reversible temperature can be either T_g or melting point T_m of the polymer.

Two-way SME: Polymer can reversibly switch between shapes without the application of external mechanical manipulation, usually due to the reversible phase transition.

Triple SME: Triple-SME utilizes the two distinct thermal transitions of a cross-linked network [55]. Those two thermal transitions are used in fixing and recovering two temporary shapes, in one shape memory cycle.

Multiple SME: Multi-SME is possible by the addition of more than two distinct thermal transitions in the materials. This can be achieved through alternation in the chemical composition of the material leading to different transition temperatures. The quadruple-SME for a multi-phase polyolefin system has been reported in the literature [56].

2. Filler-Enhanced Shape Memory Properties

The idea of adding fillers in SMPs to overcome the existing limitations is the driving force behind the designing of shape memory composites and nanocomposites. The incorporation of micro-sized or nano-sized fillers has indeed proven to improve the shape memory properties in the resulting polymeric composites. Most of the fillers such as glass fibers, Kevlar fibers, short carbon fibers, carbon nanotubes (CNTs), cellulose micro and nanofibers (CNFs), nanoclay, SiC/SiO_2 particles, etc., as fillers greatly enhance the elastic modulus, transition temperature and/or recovery stress of SMPs [15]. With their high aspect ratios and elastic modulus, nanodrods and nanofibers such as CNTs and CNFs are very effective in reinforcing and enhancing the shape recovery stress of SMPs [15]. Some of the fillers can be chemically bonded with SMP chains, thus improving the performance and mechanical properties of SMPs effectively [57,58]. Inorganic fillers such as SiC, alumina, boron nitride, and aluminium nitride (AIN) can positively affect the SME by improving the thermal conductivity and thermal stability of the polymer matrix [15]. Conductive fillers such as carbon black, CNTs, carbon nanopaper, and Ni nanorods can improve the SME in SMPs. Alternatively, different polymer fibers can also be incorporated as a filler material to enhance the shape memory properties of polymers. For example, Figure 2A shows the electrospun PVA fibers as a thermally responsive reinforcing filler to bestow a polyether block amide thermoplastic elastomer matrix with shape memory characteristics [59]. The incorporation of reinforcing 15 wt. % of CNTs in ionic elastomers based on carboxylated nitrile rubber (XNBR) showed improved shape fixation and shape recovery in progressive cycles, as shown in Figure 2B [60].

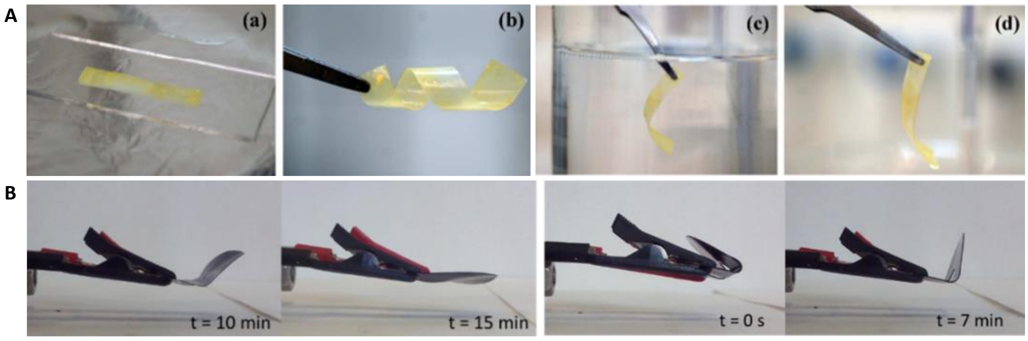

Figure 2. (**A**) Pictures showing the shape memory behavior of a polyether block amide thermoplastic elastomer with 20 wt % electrospun polyvinyl alcohol as a filler. (**a**) The sample was heated to 90 °C and formed into a spiral shape, and the temporary shape was fixed by cooling to ambient temperature (**b**). The original shape was recovered (**c**,**d**) upon heating the sample again to 90 °C in silicon oil [59]. (**B**) Electroactive shape recovery behavior of the carboxylated nitrile rubber nanocomposite with 15 phr of CNTs as a function of the time of application of a constant voltage of 50 V [60]. Reprinted with permission from [59,60].

3. Biomedical Applications of SMPs

The application fields of SMP materials are rapidly growing, and they are emerging as the foremost member of the world of shape memory materials (SMMs). Owing to their advantages such as high versatility and low cost, SMMs have a wider range of applications including deployable structures in outer space, textiles, active disassembly of electrical devices, and importantly, biomedical devices [61]. A well-known conventional use of SMPs is heat-shrinkable tubing. Industries have employed PTFE-, PVDF-, FEP- and PVC-based polymer systems for such applications and they are commercially available with a range of various diameters, transition temperatures, and other additional properties. On heating to the transition temperature, the tube shrinks, thus providing fixation/insulation/sealing/protection. SMPs and shape memory polymer composites (SMCs) have been designed, engineered, and demonstrated for a wide range of applications. The application-oriented current research regarding SMPs and shape memory cycles (SMCs) is more focused on their use in biomedical fields.

The biomedical field is an interdisciplinary field that continuously develops new materials for increased performance and functionality considering the biocompatibility of a biomaterial [62]. Polymeric materials have been explored to devise biomaterials with several functionalities [63,64]. For instance, besides a structural role, in recent years polymeric biomaterials also have biodegradability and drug release uses [62]. Owing to their SMEs, SMPs have been increasingly used for their potential in minimally invasive surgical procedures, where a compacted device is introduced in vivo through a smaller incision, and once inside the body is deployed to its shape by the suitable trigger. SMPs have been used in the design of various medical devices such as orthopedic devices, vascular stents, orthodontic wires, craniofacial mesh and plates, and neuronal probes, a few of which are discussed below. Various applications of SMPs are shown in Figure 3.

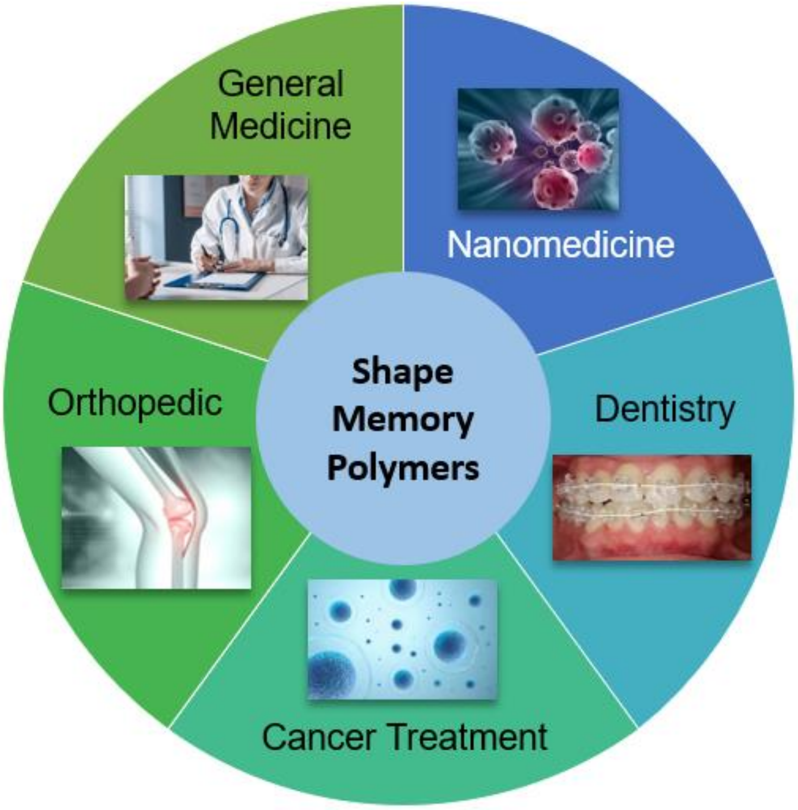

Figure 3. Biomedical applications of shape memory polymers (SMPs).

The transition temperature of bio-based SMP composite comprising poly(propylene sebacate) and embedded boehmite nanoplatelets may be brought close to body temperature, and thus increases SMC potential for biomedical applications [65]. Liu et al. showed a thermal and water-responsive potential biomaterial exhibiting T_{trans} close to human body temperature [65]. SMPNCs can be developed by cross-linking between cellulose nanocrystals (CNCs) with poly- (ethylene glycol) and polycaprolactone. The SMNC showed improved mechanical properties due to CNCs and biocompatibility on osteoblast cells. In magnetically-active SMC, the magnetic fields can fully penetrate the human body without reducing their intensity and they utilize surface-coated/modified super-paramagnetic nanoparticles such as Fe_3O_4 particles. These have wide biomedical applications. Such magnetic nanoparticles are suitable for in vivo use and can degrade in the human body harmlessly [65].

SMPs are also mixed with other polymers for modifying or tuning the SME. SMPCs incorporating lactic acid-based polymers such as polylactic acid (PLA) can be applied for biomedical applications [66]. Blending of poly(Llactide) (PLLA) with poly(ethylene vinyl acetate) (PEVA) exhibits SME [65]. The blending of PLLA into PEVA enhanced the shape fixity and shape recovery, and in addition, reduced the dependence of fixity on the fixing temperature. Moreover, the addition of PLLA in the polymer blend can improve the biodegradability and biocompatibility of the SMP blend and it can be used for biomedical applications. The SMPs can be fabricated by blending a mixture of amorphous and semicrystalline polymers. Blends can comprise a crystalline polymer such as poly(vinylidenefluoride), polylactide, poly(ethylene glycol), polyethylene, poly(vinyl/vinylidene) chloride and their copolymers, and amorphous polymers such as poly(vinyl acetate), poly methyl acrylate, poly ethyl acrylate, atactic/isotactic/syndiotactic poly methyl methacrylate and other polyacrylates [67]. These polymer blends can be used for medical devices such as stents, patches, other implants, and surgical tools in human health care [68].

SMPs can be produced by 3D printing, which is also known as material extrusion additive manufacturing [69]. 3D printing allows smart structures to be created in a wide variety of geometries and is suitable for producing SMP parts retaining good shape memory characteristics [70–72]. Different heat treatments have improved the properties of SMPs. Annealing the SMPs at 85 °C for 2 h improves the shape recovery with mechanical properties.

3.1. General Medicine

One important application of SMP is for the widely-used medical stent, which is an expandable device used to prevents vasospasms and restenosis of a blood vessel after balloon angioplasty [62]. The utilization of polymers in stent design has led to the most recent breakthrough in stent technology: drug-eluting stents. SMP stents offer the advantages of biocompatibility, biodegradability, drug loading, reduced cost, ease of fabrication, and of being minimally invasive. SMP stents can be programmed to deploy slowly at body temperature (Figure 4) and can offer improved strain recovery [73]. In 2003, Wache et al. were the first to develop the SMPU stent as a vehicle for drug delivery [74]. Gall and Yakacki et al. investigated SMP stents of thermoset (meth)acrylates by characterizing the SME [75]. This was followed by in vitro investigation of unconstrained recovery of SMP stents in 2007 [73]. To optimize the deployment of the stents, the T_g and E_r values of the polymer were independently tailored. The deformation recovery rate of the stents increases with increasing E_r and decreasing T_g.

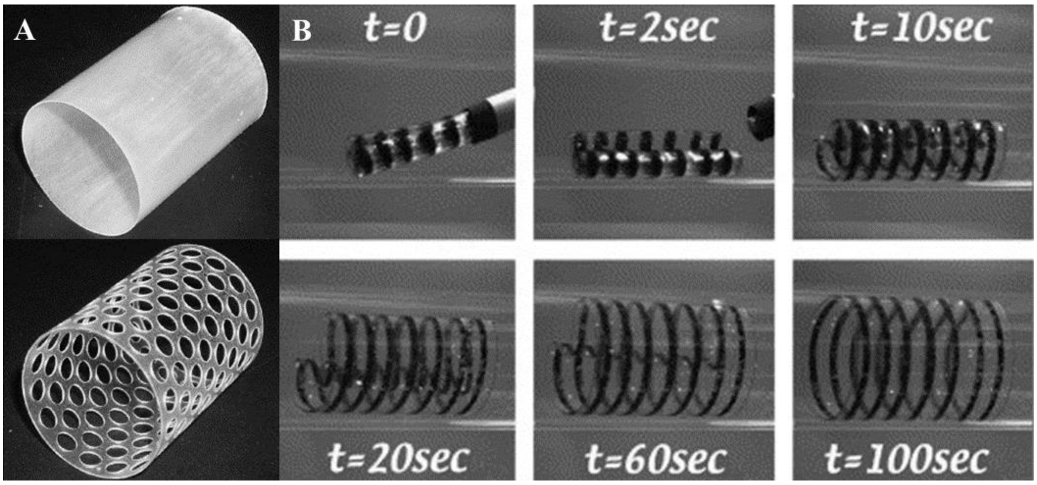

Figure 4. Photo sequence of shape memory polymer stent and its recovery. Solid and 50% perforated stents (**A**). The stent packaged in an 18 Fr. Catheter expands to 22 mm in a 37 °C water bath (**B**). Reprinted with permission from [73].

SMP foam has structural and mechanical characteristics that can be an alternative treatment in neuromedicine for intracranial aneurysms [76,77]. SMP foams have low densities, with porosities as high as 98.8%—favorable for catheter delivery and aneurysm filling, but unfavorable for attenuating X-rays. An amount of 4% by volume loading of tungsten incorporated into our SMP foams has proven to be an effective method for improving the radio-opacity of this material while maintaining the mechanical, physical, and chemical properties of the original formulation. In addition, Baer et al. [62] used the SMPs for neurovascular and laser-activated stents. In 2000, the Igaki-Tamai stent's self-expansion ability and the deployment time were reported as 0.2, 13, and 1200 s at 70, 50, and 37 °C, respectively [78].

Shape memory composites (SMCs) filled with conductive or magnetic fillers often serve to overcome limitations of SMPs and provide alternative methods of triggering shape recovery or multiple SMEs, and thus increase their potential for biomedical applications. CNT-reinforced SMPUs have potential applications in tracheal and laryngeal stents for providing mechanical strength to prevent the trachea and larynx from collapsing [65]. These devices, when inserted in the desired location, can expand using SME.

Another potential application is to curb a patient's appetite by remote inflation of an intragastric implant [65]. Thermal actuation of SMMs inserted in body might lead to the damage of nearby tissue, because of the local heating entailed during actuation. This can be avoided by using water-actuated SMCs simulating bloodstream. Such water-active SMCs are used in endovascular applications [65].

3.2. Drug Delivery

SMPs have gained interest for temporary drug-release systems that should be anchored in the body by self-sufficient active movements of the polymeric matrix [79–83]. Biodegradable polymers with shape memory effect are utilized for controlled drug release. The SMP system carrying drug could release it inside the body, through shape recovery induced by uptake of water or solvent under physiological conditions. Temperature-sensitive polystyrene and PU-based SMPs can be actuated indirectly through solvent-driven reduction of T_{trans} [62]. For instance, T_{trans} of a biodegradable amorphous polyester urethane SMP network changed from 54 °C in dry air to 36 °C in solvent medium, indicating that this material can be used as matrix in self-anchoring implants. Such implants loaded with drugs, when inserted in the body, can slowly unfold by plasticization and subsequently release drugs without any external stimuli (Figure 5) [84].

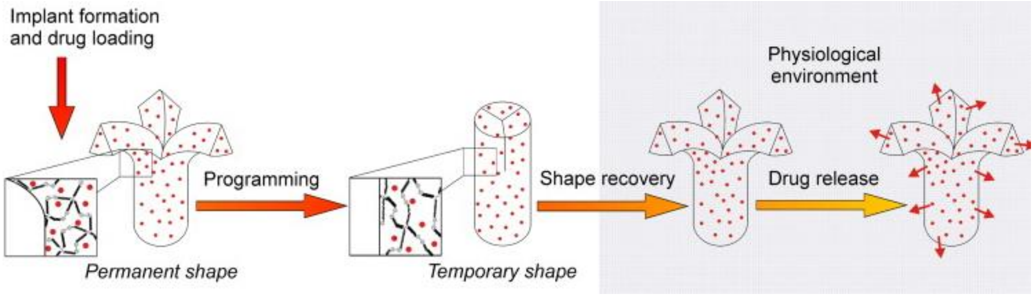

Figure 5. Programming of shape-recovery of polyester urethane shape memory polymer network in physiologic medium and temperature, and controlled drug release. Reprinted with permission from [84].

The combination of SMPs with drug release functionality results in multifunctional carriers that can be useful technology for pharmaceutical sciences [79,81]. Such technology needs molecules as (model) drugs and provides a thermomechanical characterization in an aqueous environment, reports on the loading levels on the shape-memory functionality, and explores the potential correlation of polymer degradation and drug release. This technology can be further expanded by new materials such as thermoplastic SMPs or temperature-memory polymers. However, for the application of these technologies, caution is needed to define polymer morphology as required for the shape-memory function, the processing conditions such as drug-loading methodologies, and independent control of drug release and degradation with time.

3.3. Regenerative Medicine

SMPs can be modified and have been used for wound healing and various tissue regeneration, especially for nerves [80,85]. Segmented thermoplastic polyurethanes (STPUs)-based SMPs are able to maintain their thermomechanical, chemical, and shape memory properties after the implantation and provide a reliable platform for controlled response [86,87]. STPU can be actuated at set time points to deliver bioactive agents, or cleavable compounds incorporated into a stable SMP that responds to wound signals and can be used in diagnosis [86,88]. Ramezani and Monroe et al. [86] incorporated magnetic nanoparticles into an STPU system to provide a magnetically-actuated SMP that induced 44% shape recovery only after 5 min of exposure to a magnetic field. Magnetic nanoparticles into shape memory PU systems can provide a platform for future generations of STPUs that include other cleavable or responsive components so that wound or clinician-controlled signals can act as external stimuli to cause shape and morphological changes [86,89,90].

Tohu et al. [80] developed and studied the self-expanding, watertight and biodegradable patch for fetoscopic myelomeningocele (MMC) prenatal repair. They made the tailored patch from poly (L-lactic acid) and poly (ε-caprolactone) blend and validated in vitro to accomplish all these requirements [81]. The designed patch used as a dural substitute will apparently not produce inflammation, scar formation, or tethering cord, and not induce any adverse effect on regular functions of the spinal cord. Further studies are needed to evaluate potential improvements of this novel polymeric patch in spinal cord regeneration using spina bifida models.

Similarly, Pisani et al. [85] developed the shape memory engineered scaffold, focused on potentially improving myelomeningocele (the most common type of spina bifida defect, a neural tube defect) repair in the fetus. They used copolymer poly-L-lactide-*co*-ε-caprolactone (PLA-PCL, 70:30 ratio), due to its glass transition temperature (T_g) being close to the physiologic temperature (32–42 °C). This SMP blend was used as a composite to produce electrospun scaffolds that were engineered with MMCs from amniotic fluid and the fabricated engineered scaffolds followed with favorable results.

3.4. Neuromedicine

SMPs have been also demonstrated for developing clot-removal devices. Studies reported that thermoset Pus were used for the treatment of stroke [62]. An SMP device was fabricated as a wire that punctures the clot, is then activated to coil shape, and then the clot is removed by retracting the device [91]. More recently, the Maitland group has developed a thermosetting PU system, in combination with the superelastic Nitinol wires triggered via electro-resistive heating [62].

The area of neuronal probes is an interesting opportunity where SMPs can be utilized [92]. A neuronal probe is usually a conductive electrode that can provide an electrical interface with the brain, when inserted into brain tissue. Current technologies suffer from major drawbacks, as probes employed are metallic or ceramic-based and much stiffer than brain tissue, and can cause tissue damage during rapid insertion. SMPs with their distinct advantages offer solutions to such drawbacks in neuronal probes. The softening behavior of SMPs could possibly be used to enable probe insertion in the glassy state at their transition temperature, and once implanted, the probe could be softened into a rubbery state using the local thermal energy from the brain tissue. Researchers have fabricated SMP probes and have shown trained self-insertion at slow rates causing less tissue damage [92].

3.5. Orthopedics

Common orthopedic procedures in sports medicine are repairing ligament or tendon damage that involves soft-tissue reconstructions. Yakacki et al. [93] investigated the SMPs for use in soft-tissue repairs. They showed a SMP device in 2008 that could be inserted in bone and then can expand after heating to body temperature (Figure 6) [93]. They further presented an acrylate system for such devices tailorable through variation of cross-link density and thus the recoverable force.

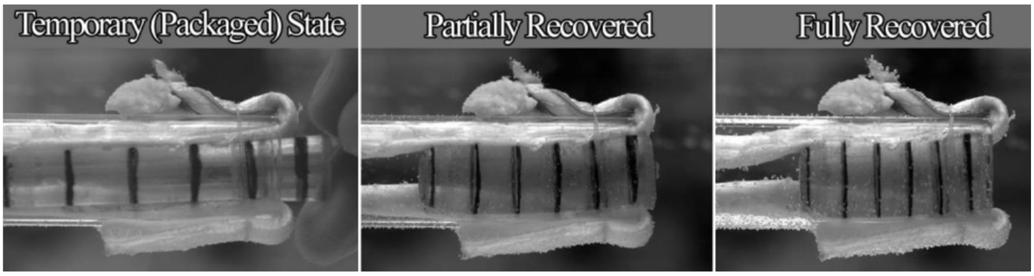

Figure 6. A shape memory polymer cylindrical that expands at body temperature. Reprinted with permission [93].

3.6. Dentistry

There is increasing use of SMPs in dentistry because of their good biocompatibility and durability in implants as well as in restoration procedures [94–96]. SMPs avoid biofilm formation during caries or root canal treatment, as well as in implants. SMPs can also be a good alternative for the metal wires in orthodontic treatments because the metallic ions are not released from the SMPs, preventing chronic diseases. Additionally, the SMPCs and SMPNC can be used for various applications, such as sustained release, root fillers, and biofilm preventative agents.

SMPs are desirable in the orthodontics area for their shape-memory effect [97]. Jung and Cho [98] in 2008 presented the use of SMPUs for archwires in a dental model. The melt spun PU, synthesized from 4,4_-methylene bis(phenylisocyanate) and polycaprolactone (PCL) diol was employed. The SMPU wires were stretched to the required length for realignment of the teeth before attaching to stainless steel brackets. Figure 7 shows the correction of misaligned teeth in orthodontics using shape memory NiTi wire and polymer-coated wire using Teflon-coated NiTi wire (Hubid Co. Ltd., Dongan-gu, Gyeonggi-do, Republic of Korea). Such wires are esthetics compared to the normal NiTi wire.

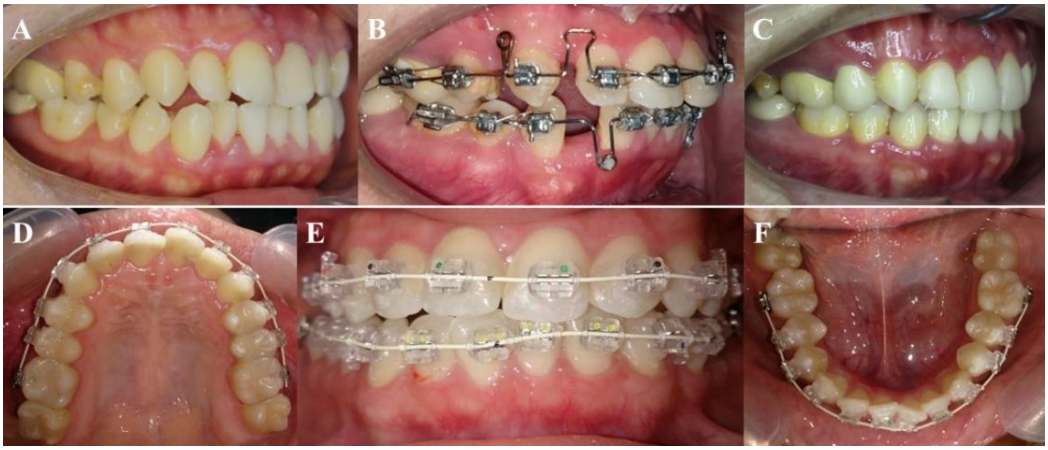

Figure 7. Use of shape memory NiTi wire and Teflon-coated NiTi wire to close the space in orthodontic treatment: (**A**) before treatment; (**B**) during treatment; (**C**) after treatment for the NiTi alloy and (**D**) before treatment; (**E**) during treatment; and (**F**) after treatment for the Teflon-coated NiTi wire.

At present, orthodontic aligners are widely used in the alignment of teeth (Figure 8). One drawback of such aligners is the change regime (7–14 days) due to the rate-limiting step. Therefore, researchers are working on improving the aligner materials, force systems, and staging of tooth movements. The incorporation of stimuli-responsive smart polymers can recover their original shape upon exposure to external stimuli [99]. This can be a great advantage in orthodontic movement.

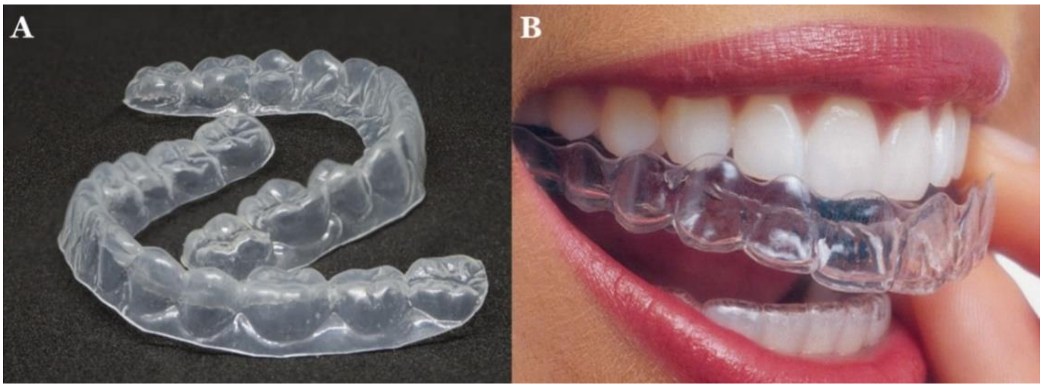

Figure 8. Aligner in the alignment of teeth. Aligners (**A**) and in mouth (**B**).

Simon et al. [99] studied the forces and moments provided by a single aligner and a series of aligners, and investigated the influence of attachments and power ridges on the force transfer. They found that initial moments were about 7.3 N·mm for maxillary incisor torque and about 1.0 N for the distalization of teeth. They concluded that apart from a few maximal initial forces, the forces and moments generated by aligners were within the range of orthodontic forces. The force change is exponential while the patient is wearing aligners due to shape recovery properties.

The success of root canal therapy depends on the cleaning and shaping, sealing of the root canal system, and microbial control. This is possible with the use of elastic shape memory endodontic instruments [100]. Biodegradable polymeric composites comprising matrixes and bioactive ceramic fillers are used extensively used in endodontics. PCL is a semi-crystalline polymer, and its properties (strength, modulus, SME, and biodegradability) depend on the crystalline fraction. Its properties have been studied for application in endodontics [101,102]. Elzubair et al. [102] studied the physical characteristics of thermoplastic synthetic degradable PCL (ResilonTM) for endodontic obturation, which contains bioactive glass and radiopaque fillers. Its performance is similar to gutta-percha in handling properties. During retreatment, they can be softened with heat (T_m is very low) and dissolved with solvents like chloroform.

3.7. Cancer Treatment

Various SMPs are used in cancer treatments. An automated head-and-neck immobilizer that can be used during radiotherapy and evaluated the positioning reproducibility in a phantom experiment [103] is an important determinant of the success of radiotherapy. Studies have been completed for applications such as magnetic resonance imaging contrast agents and hyperthermia procedures for cancer treatment [65,104,105]. Magnetically triggered SMC also provides the benefit of remote actuation of the shape memory effect. This is very useful in the biomedical field as magnetic fields are frequently employed. This technology allows medical personnel to undertake contactless mechanical adjustments in the patient's body. Magnetically active SMC could also be used for remote drug delivery for cancer.

Ouchi et al. [106] used shape-memory balloon (SMB) to improve bone cement injection efficiency and postoperative thermo/chemotherapy for bone tumors. The SMB consists of biodegradable poly(ε-caprolactone) (PCL), an anticancer drug (doxorubicin, DOX), and heat-generating magnetic nanoparticles (MNPs). This allows safe inflation at the affected site with a 400% expansion rate by simple blow molding, and in vitro cytotoxic studies showed that heat generation/drug release and only drug release from the balloon kill approximately 99% and 60% of human osteosarcoma cells, respectively. The SMB can be promising in postoperative local thermo/chemotherapy for bone tumors.

3.8. Biocompatible Protective Coating

Corrosion is an electrochemical phenomenon causing deteriotion of biomedical metals which can cause deteriotation of metal, leading to release of metal ions, which can result in tissue reactions and toxicity [107–109]. Corrosion-resistant coatings have played an essential role in corrosion protection [110]. Polymer coatings employed to prevent corrosion include alkyd and epoxy ester coatings, urethane coatings, both acrylic and epoxy polymer radiation curable coatings, vinyl, and acrylic or styrene polymer combination latex coatings [110].

Such multifunctional coatings are tailored to specific needs. Corrosion detection coatings' detection of corrosion at the right time has a direct impact on corrosion economics [110]. Recently, smart coatings have been developed which include self-healing coatings and graphene-based coatings [111]. Graphene dispersion in the polymer has led to anticorrosive properties and the advanced actuation performance coatings for biomedical applications [110,112]. Additionally, the addition of graphene has resulted in fast switching of temporary shape to original shape in polymer/graphene nanocomposites.

Some additives, known as corrosion inhibitors, can be added to polymeric coatings to aid the passive layer, such as zinc phosphate, which can assist in the formation of a phosphate layer to enhance the protective layer on the metal [113]. In addition, anticorrosive pigments can be added. They have salts which can leach out and reach the interface, and they react with metal to form a passivating layer. Additionally, oxygen scavengers can be added to the coatings. Finally, organic coatings shield the passive layer from environmental stress, so it remains intact and is not chipped off [114]. Finally, nanoparticles can be added in a little amount to form a good network in polymers to enhance the properties for biomedical applications [112,115,116].

3.9. Other Applications

Water-responsive SMPs have become significant, especially for in vivo applications, due to the intrinsic biocompatibility and ready availability of water. Guo et al. [117] designed a new water-responsive SMP using poly(butanetetrol fumarate) (PBF) and its properties were modulated by curing. The PBF scaffolds exhibited high shape recovery and fixity rates (>95%) and showed good biodegradability, and it could support the attachment, viability and alkaline phosphatase activity of osteoblasts. The PBF SMPs can be used as water-responsive scaffolds, sensors or actuators. Furthermore, PBF could be readily functionalized via pendant hydroxyl groups, which was demonstrated by the immobilization and controlled release of bone morphogenetic protein 2 (BMP-2). Recent studies on SMP-based conductive composites using CNTs show potential applications as electroactive and remote-sensing actuators [27]. Furthermore, SMPs have also been tried in the making of artificial muscles to replace lost muscle in humans [118]. However, these need more clinical studies.

4. Shape Memory Composites (SMCs)

To overcome the limitations of SMPs, fundamental research on the fabrication of SMP composites has been in high demand to meet the requirements of various applications. The addition of small amounts of micro/nano-sized fillers into SMPs can significantly improve the material's thermo-mechanical properties, and enhance the shape-memory effects and processability. Fabrication of SMCs has enabled engineering technology to combine the filler properties with polymer chemistry to achieve SMMs. The micro/nano-sized fillers provide control at the molecular level over polymer dynamics, surface/bulk properties, and their functions. The interfacial interaction between the polymer matrix and embedded fillers has enabled the adjustment of the stiffness and elasticity of the composites. The type of filler, amount, and surface modification give control over these interfacial interactions with the matrix to get the particular properties in SMCs. SMP composites filled with particles or fibers have been studied comprehensively to enhance the mechanical properties, SME, and develop more functions in SMCs. Some novel fillers such as CNFs, CNTs, nanoclay, graphite, selenium, and graphene, have resulted in SMPs with high performance and tailored properties for desired applications. Owing to their outstanding properties such as low cost, light weight, processibility, and high strain recovery, SMCs make versatile candidates for various potential applications. However, although many studies regarding the potential applications of SMCs have been reported, many of these reports only demonstrate possible applications of the SMCs. The practical applications of SMCs are still limited, as they require a more specific set of properties in SMCs. For instance, SMCs in textile applications may have good shape fixity, but are less elastic. In the case of biomedical applications, SMCs are required to meet several parameters including high stiffness, recovery strain/stress, and safe stimulation method. SMCs should be evaluated for their biocompatibility, biodegradability, and degradation products. For example, CNT/SMP composites have to be evaluated stringently before biomedical applications, as MWCNTs have high toxicity and hydrophobicity [119]. Key issues such as the thermal and chemical stability of the SMCs should be taken into account for their applications. The SMCs may be subjected to sterilizations prior to use in medical applications and the effect of sterilizations on the properties of the material should be systematically investigated. Although in the past, researchers have invested great efforts to overcome many of these problems, SMC products still face challenges that need to be addressed in advance of their vast growth. One way to increase the potential applications of SMCs in the medical field is the bio-based matrix, and fillers could be employed in the fabrication of the composites. The commercially-available sources of the matrix, appropriate biocompatible fillers, and more technologically viable fabrication processes would be the key steps to achieve the practical applications of SMCs. Furthermore, an alternative fabrication approach for SMCs that utilizes the combination of commercially-available elastic polymer matrix and a micro- or nano-sized filler to achieve shape memory as an emergent property in resultant composites is attractive. SMPBs have enabled a combination of the two existing polymers with or without intrinsic SM properties to achieve shape memory behavior in resulting polymer blends. Such polymer blends gives a platform to tailor the SM properties of existing SMPs or to formulate new SMMs from non-SM-based polymers. The SM polymer blends have provided a platform for the fabrication of SMMs from commercially-available sources at a technologically viable scale. However, these SMPBs were more recently developed, and they need to be further investigated for their design, hermos-mechanical properties, and shape memory properties. They also need to be demonstrated for their potential applications in various fields, especially the biomedical field.

The field of SMP materials has seen a tremendous amount of growth and development since its first discovery. Molecular architecture and an underlying mechanism enabling SME in polymers have been well-studied and -classified. SMPs have been categorized based on the type of stimulus used for their shape memory actuation. An important motivation for rapid growth in the field of SMPs is their broad application potential. The potential applications of these SMPs can be found in everyday life [62]. This has extended application fields to wider operations in the area of medical devices, such as implant dentistry, prosthetic dentistry, and orthodontics [62]. The fundamental research is now focusing on new types of SMPs and the mechanisms of SME. Volumetric additive manufacturing is an emerging paradigm that can generate complex 3D SMP structures without layers [120].

The relatively low mechanical properties are still limiting the application of SMPs in various fields. In addition, detailed studies of structural phenomena of SMPs that affect shape memory properties, such as fatigue, relaxation, creep, and duration, need to be carried out. The significant SMP research is driven by their tremendous scope in biomedical applications. However, stringent biological evaluation of SMPs is important prior to their applications as biomaterials. The typical triggers of SMP such as heat, UV light, and electricity may be harmful to humans [117]. The implementation of a safe and effective thermal actuation of SMP devices in vivo is one of the key challenges in their application in the medical field.

5. Conclusions

It is evident that SMP materials have expanded their application areas in the field of biomedical applications, including general medicine, drug delivery, regenerative dentistry, dentistry, neuromedicine, cancer therapy, orthopedics, and corrosion protection. At the same time, progress in SMP architectures and shape memory mechanisms has advanced. Extensive work has been conducted in understanding the material functionality relevant to the design, biological response, and appropriateness for end biological applications. Owing to their advantages such as high versatility and low cost, easy processability and tailored structure, further advancement for practical application areas toward personalized medicine and customized devices can be expected. Existing challenges relate to a detailed understanding of in vivo studies of these SMPs and the need for extensive preclinical investigation to translate them from lab-scale to biomedical applications.

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