An Extensive Review of Shape Memory Polymers for Biomedical Applications

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Abstract Shape memory polymer is usually referred to as SMP may be a smart polymer which may take a short lived change from its original shape with the acceptable glass transition temperature and are available back to its original state. This article focuses on the properties of various memory polymer which supports the parameters like Shape memory effect, electro conductivity, stimuli, etc. The aim of this article is to survey a number of the SMP’s viz. Polyurethane, PMMA [poly(methyl methacrylate)] and Hybrid Shape memory polymers. Since, the Shape memory polymer are often used for various medical uses where the longer term use of the acceptable electro conductive shape memory polymer is to synthesis a facial mask for the appliance of Facial reconstruction. Overall perspective of this review is to prove the best suited polymer for the utilization in shape memory polymer.

Keywords- Shape Memory Polymer (SMP), Polyurethane, PMMA, Hybrid Shape Memory Polymer

1. Introduction

Shape memory polymer is a smart material which can alter its shapes depending upon the inducing stimuli [1]. It is commonly referred to as SMP. They are also known as smart polymer materials. The property of changing shape requires a stimulus to induce the shape change hence it can be heat, light, or chemo responsive [2]. The shape memory polymer usually changes when the threshold temperature is reached that is the glass transition temperature (Tg) [3], the polymer again deforms itself with a particular stimuli. This property of the shape memory polymer is due to the polymer morphology and functionality [4]. The period where the shape memory polymer changes its form is call the shape memory effect. Based on this shape memory effect the shape memory is classified into dual shaped, triple shape memory polymer and also multiple shape memory polymers [5]. There are several shape memory polymer combinations that are being used however; the most commonly used shape memory polymer is polyurethane [6]. Other shape memory polymers that we are to discuss here are PMMA [poly (methyl methacrylate)] and hybrid shape memory polymer [7]. The aim of the project is to compare the memory polymers like polyurethane, PMMA [poly (methyl methacrylate)] and hybrid shape memory polymers. Hence this analysis helps us to spot the higher shape memory polymer for the longer term development of facial reconstruction mask which may be a facial mas to research the damages occurred within the face internally. Therefore, the polymer’s properties
are to be studied based on the various literature surveys. SMP with various analysis is done to understand properties of Polyurethane, PMMA [Poly(methyl-methacrylate)], Hybrid shape memory polymer and its merit factor.

2. Analysis

2.1 Shape memory polymer

The shape-memory function can be generated as a reversible effect enabling actuation behaviour which is known as the period of transformation with the help of an external stimuli through macro scale deformation and processing, specifically by detecting the macro molecular commencement of actuating units and of the basic structure of geometry to determine units of the polymers [8].

![Figure 1](image)

**Figure 1** Mechanism if shape memory polymer

Figure 1 explains the mechanism of the shape memory polymer where when the polymer is induced with a stimuli it programs itself into a required shape and remains in a temporary state unless the temperature variation. The shape recovery is obtained at the end during the recovery stage. It is also to be noted that above the glass transition temperature (Tg) the shape memory polymer attains a rubbery state. This state is normally induced by the stimuli, where it is deformed to a temporary state. Once when the glass transition temperature falls or the inducing stimuli is taken off as affecting the Tg the shape memory polymer comes back to its original shape[9]. It is to note that the basic important parameters of shape memory polymer are glass transition temperature [Tg], melting temperature [Tm], shape recovery ratio [Rt], shape fixity [Rf] from the Figure 1, Whereas, recovery and actuation properties are re-programmable. This reversible effect of shape memory polymer actuation is an emerging research that has a lot of emerging applications. The shape memory polymer is normally preferred because of its highly compatible nature and a light weight material [10]. Shape memory polymers with soft actuation have also attracted tons of attention within the biomedical device industry 0usch as artificial muscle fabrication.

2.2 Polyurethane

Polyurethane is a type of shape memory polymer. The glass transition temperature of the polyurethane may vary depending upon the isocyanates used however the average Tg of polyurethane may range from 210K to 310K [11]. It is basically synthesized by poly addition reaction of polyols, isocyanate, and chain extenders. Polyurethanes are in the class called reaction polymers, which also includes epoxies, unsaturated polyesters, and phenolics [12]. They are produced by the reaction of an isocyanate groups per molecule such as \((R−(N=C=O)n\) with a polyl containing hydroxyl groups per molecule \((R′−(OH)n\) with the presence of a catalyst [13].
The Figure 2 explains that the polyurethane preparation is done by the chain reaction of polyol, isocyanate [14]. The strength of the polyurethane depends upon the structure containing the isocyanates and the catalyst. The catalyst helps in speeding up the reaction and the ability of shape memory properties. The isocyanates are of various types that are used for the production of polyurethane [15]. They can be toluene di isocyanate [TDI], methylene diphenyl diisocyanate [MDI].

The isocyanate groups can be aliphatic as well as aromatic. The Figure 3 shows the methylene diphenyl diisocyanate pure form which is formed to polymeric MDI where it will be reacted in the presence of polyol and catalyst for the formation of polyurethane [16]. The properties of polyurethane are greatly influenced by the kinds of isocyanates whereas; the polyol helps in the flexible segments which helps it to maintain its elasticity. Sometimes, the high amount of cross linking may cause rigidity [17]. Long chains and low cross linking provides a polymer that’s very stretchy, short chains with many cross link produce a tough polymer while long chains and intermediate cross linking provides a polymer useful for creating foam. The cross linking that is present in the polyurethanes explains that the polymer consists of a three-dimensional network which causes the relative molecular mass to be high [18]. In some aspect, a small amount of polyurethane are generally considered as one giant molecule and the isocyanates as well as polyols, include other additives and processing conditions which allows polyurethane to possess the very wide selection of properties that make them such widely used polymers [19]. The applications of polyurethane has spread to a wide range in medical field. Due to its higher bio compatibility and elasticity during Tg has allowed it to attain various medical field applications such as catheters, removable stent and also sutures that can deform itself to a required shape [20].

2.3 PMMA [Poly (methyl-methacrylate)]

PMMA is expanded as [Poly (methyl-methacrylate)] which is an alternative to polycarbonate (PC) where the durability, flexible strength, transparency, lustrous nature, and UV tolerance are more important than impact strength, chemical resistance, and warmth resistance [21]. PMMA is synthesised by various methods of polymerization such as emulsion polymerization, solution polymerization, bulk polymerization where a large quantity of petrol is required [22].
The Figure 4 represents the structure of the poly (methyl-methacrylate) where the hydrogen and carbon are combined to form the structure [23-24]. Additionally, PMMA doesn’t contain the doubtless harmful bisphenol-A sub units found in polycarbonate and may be a much better choice for laser cutting, it is often preferred due to its moderate properties, easy handling and processing, and low cost [25]. Non-modified PMMA behaves during a brittle manner when under load, especially under an impression force, and is more susceptible to scratching than conventional inorganic glass, but modified PMMA is usually ready to achieve high scratch and impact resistance [26]. It also has good impact strength, above both glass and polystyrene; however, PMMA’s impact strength remains significantly less than polycarbonate and a few engineered polymers. The glass transition temperature of the PMMA ranges from 85 to 165 degree Celsius whereas the melting point of PMMA is 160 degree Celsius [27]. It filters most of the ultraviolet (UV) light at wavelengths below about 300 nm. PMMA also swells and dissolves in most of the organic solvents, poor resistance to several other chemicals. Nevertheless, the environmental stability is superior comparatively to most of the other plastics like polystyrene and polyethylene, and PMMA is therefore often the fabric of choice for outdoor applications [28]. However, from the survey PMMA is not as elastic as polyurethane and also doesn’t withstand the glass transition temperature where the temporary shape is formed. This makes the PMMA a bit brittle as SMP.

2.4 Hybrid shape memory polymer

Hybrid shape memory polymers are a kind of shape memory polymers where a polymer is combined with an external hybrid material so on re-inforce the properties of the form memory polymer [29]. Nowadays the hybrid shape memory polymers are participating in most of the textile, and plastic industry. It is yet to play part in medical field applications. Some of the hybrid shape memory polymer examples are Silica-polyurethane SMP, PEG-CNT (Poly ethylene glycol-Carbon nano tube) SMP, and PVC/PU (Poly vinyl Chloride-Poly urethane [30]. The transition temperature of the hybrid shape memory polymer depends upon the polymer that has been used and the combination plasticizing agent and fillers. Shape memory alloys exhibit plastic to elastic transformation upon heating, allowing the alloy to return to its original shape, and in spite of that, a rise in modulus normally follows increasing temperature [31]. The modulus is often varied three to fourfold between martensitic and austenitic phases. However, shape memory polymers believe the glass transition temperature or melting temperature of sentimental segments, and thus, unlike the metallic materials, they exhibit a change in modulus with rising temperature [32]. Shape memory polyurethane hybrids incorporating silica obtained from tetraethoxysilane (TEOS) as reinforcement were employed to enhance their mechanical properties, the form memory effect and mechanical properties of hybrids are investigated as a function of TEOS and PU hard segment [33].

The elongation-at-break increases initially with TEOS content similarly as in breaking stress, because the elongation-at-break of composites usually decreases with increasing the reinforcement [34]. it’s going to be ascribed to the deformation mechanism of PU, just in case of PU composed of hard and soft segment domain, the hard segment lamellae sensitive to applied stress are often tilted toward stretching direction within the low strain, and within the high strain, they align parallel to the stretching direction [35]. Therefore, polyurethane can maintain the stress capacity bordered in the relatively high strain with seldom breakdown of amorphous soft chains. For instance, the polylactic acid and thermo plastic polyurethane is a type of hybrid shape memory polymer. To test its stability it is combined with PEG[poly ethylene glycol] which
acts as an inhibitor for the polymer chain. That is it helps to control the displacement of the polymer chain. It is also known as the plasticizing agent. For the conductivity purpose Single Wall carbon nano tubes are used as fillers [36]. This hybrid shape memory polymer enhanced a higher shape memory performance yet not mostly used because of its immiscible blends and complicated fabrication. If the plasticizing agent is not added in a proper ratio it might affect the mechanical brittleness of the polymer. Due to the significant property of the SHP it’s used in many biomedical applications based on the biocompatibility of the materials such as nontoxic nature, compatibility of the blood and many more. Table 1 shows the nature of the material with the young modulus value and its suitability for the particular part of the human body.

| Tissue/material                        | Modulus [MPa]       |
|---------------------------------------|---------------------|
| Human tissue                          | 2x10³ – 20.4x10³    |
| Femur                                 | 2 X10³–19.3 X10³    |
| Breast tissue                         | 3.25 X 10⁻³         |
| Cornea                                | 24.5 X 10³ – 20     |
| Lens                                  | 1.1 X 10⁻³ – 10.6 X 10⁻³ |
| Tendon                                | 1.2 X 10³           |
| Skin                                  | 0.42–0.85           |
| Spinal cord & gray matter             | 2                   |
| Muscle                                | 1.45–20             |

**Shape-memory polymers**

| Polyurethanes                          |                       |
|---------------------------------------|-----------------------|
| Star-shaped cooligoesters-urethane networks of lactide and glycolide | 330 – 600             |
| Oligo(ε-caprolactone) diols with oligo (p-dioxanone) diols and diisocyanate | 34–90                 |
| Poly(ε-caprolactone) oligomers polyurethanes with N,N-bis (2-hydroxyethyl) cinnamamide | 5.01–7.26             |

| Polyesters                            |                       |
|---------------------------------------|-----------------------|
| Oligo-(ε-caprolactone) dimethacrylate and n-butyl acrylate | 0.5–71                |
| Poly(ε-caprolactone) dimethacrylate | 260                   |
| Methacrylated Poly(ε-caprolactone)   | 0.3–3.5               |
| Polylactic acid–polyglycolic acid dimethacrylate | 12–288                |
| AB-polymer networks with co-oligoester and poly(n-butyl acrylate) | 12.5                  |
| Electrospun poly(p-dioxane) and poly(ε-caprolactone) networks | 3.3–5.0               |
| Poly(trimethylene carbonate-co-D,L-lactide) random copolymers | 2.0–4.7               |
| Poly(ε-caprolactone) networks crosslinked with cinnamoyl moieties | 223–511               |

| Biopolymers                           |                       |
|---------------------------------------|-----------------------|
| Chitosan                              | 4246                  |
| Poly[(3-hydroxybutyrate)-co-(3-hydroxyvalerate)] | 100.1                |

| Other polymers                        |                       |
|---------------------------------------|-----------------------|
| Poly(glycerol dodecanooate)           | 136.55                |
| Acrylate-methacrylate networks constituting ethylene glycol phenyl ether acrylate and ethylene glycol phenyl ether methacrylate | 0.7–31.2              |
| Methylmethacrylate-co-polyethyleneglycolmethacrylate | 9.3–23                |
| 2-(dimethylamino) ethyl methacrylate, butyl acrylate and tri(ethylene glycol) divinyl ether copolymers | 60–625                |

Source: Table 1 is depicted from Adv. Funct. Mater. 2020, 1909047
3. Biomedical Applications

SMP is used in various biomedical application with single polymer and as polymer matrix as listed in the Table 2. Some of the promising biomedical applications of the SMP are as listed [37].

- Drug delivery
- Cardiovascular applications
- Tissue Engineering
- Antibacterial applications
- Wound dressing
- Stent

| Chemical composition | Trigger | Form       | Application        | Notable characteristics                                      |
|----------------------|---------|------------|--------------------|----------------------------------------------------------------|
| Co polyester urethane with oligo[(rue-lactide)-co-glycolide] | Thermal | 2D films   | Drug delivery      | Tests different drugs                                          |
| PLA/PCL-based poly(urethane) + iron oxide NP + PEG/gelatin | Thermal/moisture | Scaffold   | Bone tissue engineering | Biodegradable                                                  |
| Polyurethane          | Ultrasound | 2D shapes | Drug delivery      | Biodegradable                                                  |
| PCL-based polyurethane | Thermal | Tubes      | Vascular graft     | Biocompatible                                                  |
| L-lysine               | Films    | Sutures    | Sutures            | Biodegradable                                                  |
| Oligo(e-caprolactone) diol + oligo(pdioxanone) diol + diisocyanate | Thermal | Monofilament | In vivo study in a dog | Biodegradable                                                  |
| Poly(urethane) + CNT  | Thermal | Foam       | Intravascular treatment of | Recovery through resistive heating |
| Poly(urethane)        | Photothermally | Foam | Aneurysm occlusion | Laser activated                                                  |
| Poly(urethane)        | Photothermally | Stent + foam | Vascular stent | Laser activated                                                  |
| Poly(urethane) + NiTi | Electro resistive heating | Microactuator coil | Clot removal | In vitro model                                                   |
| Poly(urethane)        | Photothermally | Microactuator coil | Clot removal | Laser activated                                                  |
| Poly(urethane)        | Thermal | Spiral fiber | Nerve stimulation | In vivo studies in mice                                         |
| Poly(carbonate urethane) + Fe3O4 microsphere | Thermal | 2D films | Cerebral embolization | Decreased platelet adhesion, Promotion of rat vascular smooth muscle cells |
| Poly(e-caprolactone-co-DL-lactide) | Thermal | Tubular stent | Esophageal stenosis | Biodegradable                                                     |
| Methacrylated PCL     | Thermal | Tubular    | Airway stent       | SLA printing                                                      |
| Material Description                                      | Manufacturing Method | Product Type | Application                          | Additional Information                                           |
|-----------------------------------------------------------|----------------------|--------------|--------------------------------------|------------------------------------------------------------------|
| Poly(ε-caprolactone) + hydroxyapatite nanoparticles       | Thermal              | 2D films     | Bone defect repair                   | Presence of hydroxyapatite nanoparticles                          |
| Hydroxypropyl                                             | Thermal              | 2D films     | Drug delivery                        |                                                                  |
| Oligo[ε-caprolactone-co-glycolide]                        | Thermal              | 2D films     | Drug delivery                        |                                                                  |
| Poly(D,L-lactide-co-trimethylene carbonate)               | Thermal              | Monofilament | Sutures                             |                                                                  |
| Oligo (ε-caprolactone-co-glycolide)                       | Thermal              | Ureter stent | Ureter stent                         | In vitro model                                                   |
| Dimethacrylates                                           |                      |              |                                      |                                                                  |
| Poly(ε-caprolactone) + silver                             | Thermal              | 2D films     | Self-tightening sutures              | Antibacterial                                                    |
| Ureidopyrimidinone modified gelatin                      | Thermal              | 2D films     | Drug delivery                        | ECM mimic                                                        |
| Chitosan + carbon nanotubes (CNT)                        | Hydration            | Foam         | Noncompressible hemorrhage           | In vivo study in mice and rabbit                                 |
| Chitosan, poly(ethylene oxide), glycerol                 | Hydration            | Helical stent | Vascular stent                       | In vivo study in rabbits                                          |
| PVA-based                                                 |                      |              |                                      | Biodegradable                                                    |
| PVA + glycerol                                            | Hydration            | 2D films     | Drug delivery                        | Additive manufacturing                                           |
| Gluteraldehyde crosslinked                               | Hydration            | Circular staple | Anaestomosis of hollow              |                                                                  |
| Poly(vinylpyrrolidone-co-acryloxyacetophenone) organogels | Thermal              | 2D films     | Limb support structures              | Room temperature formation of temporo-                           |
| Poly(acrylonitrile-co-acrylamide) + PEG dimethacrylate + BaSO₄ | Thermal              | Microcoils   | Embolization of arteries             | Radiopaque                                                       |
| Poly(octamethylene maleate (anhydride) citrate)          | Thermal              | Scaffold     | Cardiac tissue patch                 | Minimally invasive surgery                                       |
| Poly(N-acryloyl glycaminide) + nanoclay                  | Thermal              | Scaffold     | Bone tissue engineering              | Additive manufacturing                                           |
| Poly(ethylene glycol phenyl ether methacrylate)           | Thermal              | 2D films     | Ophthalmic devices                  | Optical characterization                                          |
| Poly(glycerol sebacate) + poly(1,3-propylene sebacate) + kartogenin | Thermal              | 2D films     | Cartilage regeneration              | In vivo study in rats Cell-free regeneration                      |
| Poly(butylnmethacrylate-co-methacrylate)                 | Ultrasound           | 2D shapes    | Drug delivery                        | Localized recovery/release                                       |
|                                                          |                      |              |                                      |                                                                  |
|                                                          |                      |              |                                      | Temporal control over recovery/release                            |

**Note:** The table includes a variety of materials used in different biomedical applications, such as bone defect repair, drug delivery, and tissue engineering. Each entry specifies the material type, the manufacturing method (Thermal, Hydration, Photothermal, etc.), the product type (films, foam, etc.), and the specific application (bone defect repair, drug delivery, etc.). Some entries also note additional features such as presence of hydroxyapatite nanoparticles, drug incorporation methods, and in vivo degradation and other studies.
Drug Delivery is becoming a very interesting and significant area of research in the recent years and targeting a particular sigh has been very important to cure diseases like cancer. SMP have been used in the drug delivery applications which has a better reflex on the reorientation to its original shape based on the physical parameters such as change in temperature. Research has been carried out with copolyester urethane to release the drug in aqueous environment [38]. Similar to the drug delivery applications the SMP are used in the field of cardiovascular applications as intravascular stents for blocked or narrowed artery [39]. Antibacterial applications of the SMP were analysed with the Diketopyrrolopyrrole - based conjugated polymer as photothermal filler in a polycaprolactone-co-polyurethane matrix which had antibacterial effect on irradiation with the infra-red effect to recover its original shape [40]. SMP are used in similar other applications as stated on the tables 1 and 2 based on its nature and compatibility for the particular biomedical applications.

4. Inference

- From the analysis of the various shape memory polymer discussed in this article, we hereby infer that polyurethane has the higher eligibility for the futuristic development of facial reconstruction mask.
- Polyurethane has a unique characteristic because of its amorphous and crystalline phase due to which the incompatibility occurs due to the unequal homogenized chain molecules.
- The PMMA is mostly of a programmable chain of shape memory property which has tiny influence of shape memory effect.
- This PMMA is the commercial engineering polymer which has a very low recovery rate compared to that of other shape memory polymers, it also has a disadvantage of the material becoming hard and less ductile when kept in low temperatures.
- When the polyurethane was combined with the silica clay micro particles it showed enhanced changes in the colour as well as thermal properties [41]. It also showed very less recovery state and low compatibility.
- The PMMA is also combined with titania as a hybrid shape memory polymer which helps to improve the flexibility and also toughness of the polymer. But it has a disadvantage of reduced recovery rate of the shape memory polymer.[41]
- Since the polyurethane is capable of self-healing it is also used as a wound dressing which can be a major advantage to use it in medical field. It is most suitable for the healing of cutaneous skin wound healing as bioactive wound dressings. It is specified that the polyurethane even when greatly damaged it takes nearly 40s to self-heal. It possesses a variety of temperature for shape recovery, high recoverable strain, inherent soft-hard segments, high control on the softening and retraction temperatures with biophysical properties [42]
- The optical properties of the polyurethane is reversible act as they could be tailored into an optical structure and also recover back to its original shape in the recovery process [43]. For the electrical property of the polyurethane it is mandatory to fill with electro active fillers such as carbon nano tubes, conductive metallic powders etc., which enhances the electrical properties of PUSMP [44].
- The higher the conductivity the more the joule heating which causes the temperature rise and speedy recovery at the specified voltage [45]. The polyurethane has also magnetic properties which act as the actuators in the shape memory effect by inducing the heat energy [46-50]. It is noted that the polymer blend of polyurethane with fillers for the various properties might have slight low fall in the property of SMP.
5. Conclusion

Overall, this project comprises of various varieties of shape memory polymer. From the inference of the papers the wide range of shape memory polymer which is playing a major role in the applications of different kind of industries has different properties based on their composition were analysed. The composition of the polyurethane is strong and has a threshold requirements with enough thermo mechanical and transition properties whereas, PMMA[poly(methyl-methacrylate)] also has wide range of properties but the elasticity of PMMA doesn’t match the polyurethane. Since PMMA is mostly used in the textile and ceramic industries it also has very feeble bio compatibility. The polyurethane also has a wider range of shape memory properties even when combined with other components to enhance the hybrid shape memory polymer properties. Thus, by the inference we conclude that the polyurethane is the best suited for component for Shape Memory Polymer (SMP).

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