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A REVIEW OF RECENT ADVANCES IN POLYMERIC SHAPE MEMORY HYDROGEL

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ABSTRACT: In Today's world, there are many drug delivery systems available but there are many problems related to drug delivery systems like stability, degradation *in-vivo*. Drug delivery through tablets is the most convenient method due to patient compliance. Recently, hydrogels are hydrophilic 3-dimensional polymeric structures that have been explored by researchers due to their extraordinary promising potential in a wide range of applications such as drug delivery, sealing, and tissue engineering. These 3-D polymer networks with the ability of shape deformations show a volume phase transition. The polymers used in the formulation of hydrogel have low mechanical strength and difficult to handle. Shape memory hydrogels have been extensively explored and probed; however, no amalgamated information on their effective utilization in biomedical applications is available as such. In this review article, we systematically consolidate prevailing literature and briefly enumerate shape memory hydrogels based on their principles as well as their shape transitions in diverse environments, which significantly affects operational exploitations of shape memory hydrogels.

INTRODUCTION: Hydrogels are water-swollen three-dimensional polymeric systems that researchers have been investigating in a wide scope of utilizations, for example, medicate conveyance, fixing, tissue building, and so forth, because of their remarkably encouraging potential. Hydrogels with a lot of water are delicate and flexible enough to permit distortion contrasted with the relating mass polymer films [1, 2]. Hydrogels are fragile three-dimensional frameworks formed by physical and additionally synthetic cross interfacing of hydrophilic polymers, which can expand holding and holding a critical whole of water.

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Hydrogel has been objective considered because of their significant properties, for example, their closeness to body tissue, low surface pounding, ability to epitomize and release (atoms, particles, and cells), fitting morphology for cell augmentation and upgrades responsive properties [3, 4].

Especially, boosts responsive practices roused essentially, for example, the opening and shutting of blooms and the arrival of seeds from pine cones are influenced by light or temperature, and are among the most alluring highlights of hydrogels that have increased exceptional consideration. For the most part, hydrogels with "keen" practices experience reversible shape miss-happenings in light of outside improvements, for example, warmth, light, or power. In this manner, a fascinating way to deal with misuse of the maximum capacity of hydrogels is to advance their self-healing and shape memory properties, which are notable for customary materials or even

polymers however these days keeps being a difficult issue for hydrogels. Self-recuperating is one of the most intriguing capacities experienced in nature, and it speaks to the capacity of a material to recover and fix itself after harm. This property depends on the reversible compound or physical communications that rapidly and proficiently are transformed inside the structure of the hydrogels, and they would permit to expanding of the working life expectancy and application edge of hydrogels. These days, as two of the most well known "brilliant" hydrogels, shape-memory hydrogels (SMHs) consideration because of their promising potential applications in biomedicine and delicate mechanical technology, and as fake muscles or as small scale swimmers. The size of hydrogel matters. Hydrogels can be thrown or framed into essentially any shape and size, as indicated by the prerequisites of. Hydrogel delivery systems can be classified into three main categories based on their size: macroscopic hydrogels, microgels, and nanogels **Fig. 1**. Microgels and nanogels are particulate hydrogels with dimensions on the order of micrometers and nanometers, respectively.Shape memory polymers (SMPs) are a developing kind of keen polymers that can change their shapes in predefined routes in light of reasonable stimulation [5-7]. Various sorts of external stimuli are used for such as trade-in temperature, mild induction, electrical current, solvents, or ion strength. One of the most frequent stimuli to set an alternate in shape is temperature as it permits convenient programming of SMP materials. SMPs can be comprised of thermoplastics with physically caused crosslinks (TP-SMP) and thermosetting polymers with covalent crosslinks (TS-SMP) as long as the molecular network shape carries at least two separate phases [8-9].

SMPs generally consist of crosslinked segments that determine the permanent shape and switching segments at transition temperatures that repair the temporary shape. The permanent structure of the SMP is decided either by bodily or chemical crosslinks. Therefore, based on the nature of the crosslinks conventional SMPs relying on thermal segment changes can be labeled into two types, these that are either chemically or bodily crosslinked. By heating the polymer above the switch transition temperature, the SMP recovers its permanent shape as a result of releasing internal stress stored in the crosslinking structure. As heat is not a convenient stimulus in practical biomedical and textiles applications, reversible interactions, such as metal-ligand coordination, host-guest interactions, and dynamic covalent bonds, have been applied to realize shape memory behavior in mild conditions [10-12].

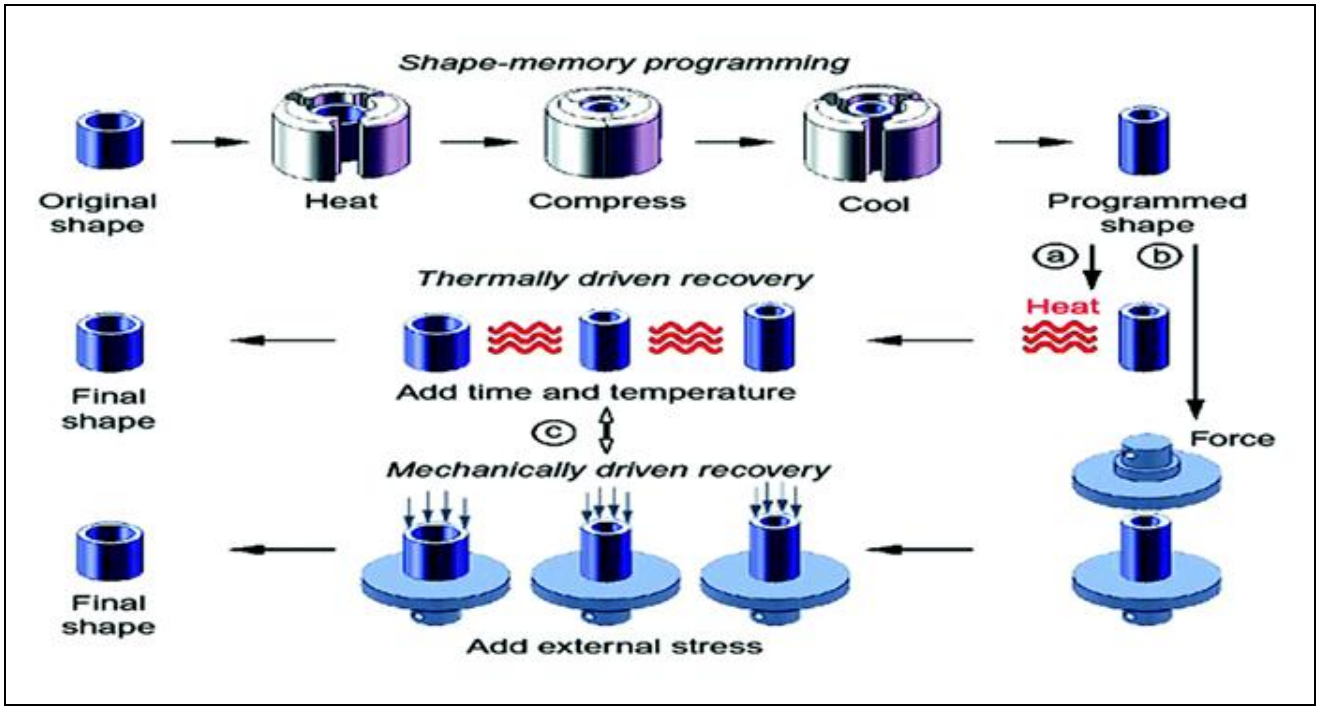


FIG. 1: MAIN STAGES OF THERMALLY INDUCED SMPS COMPARING THERMOSETTING (TS) AND THERMOPLASTIC (TP) BASED SMPS

Advantages: [13-15]

1. They can use Diverse External Stimuli and Triggers: in addition to heating, there are many alternative methods to set off the shape recuperation (*e.g.*, light, magnetic field, chemical, and power), which can coincide, prompting multi-delicate materials.

2. They Exhibit Exceptionally Flexible Programming: The programming can be achieved with specific stimuli through single and multi-step strategies.

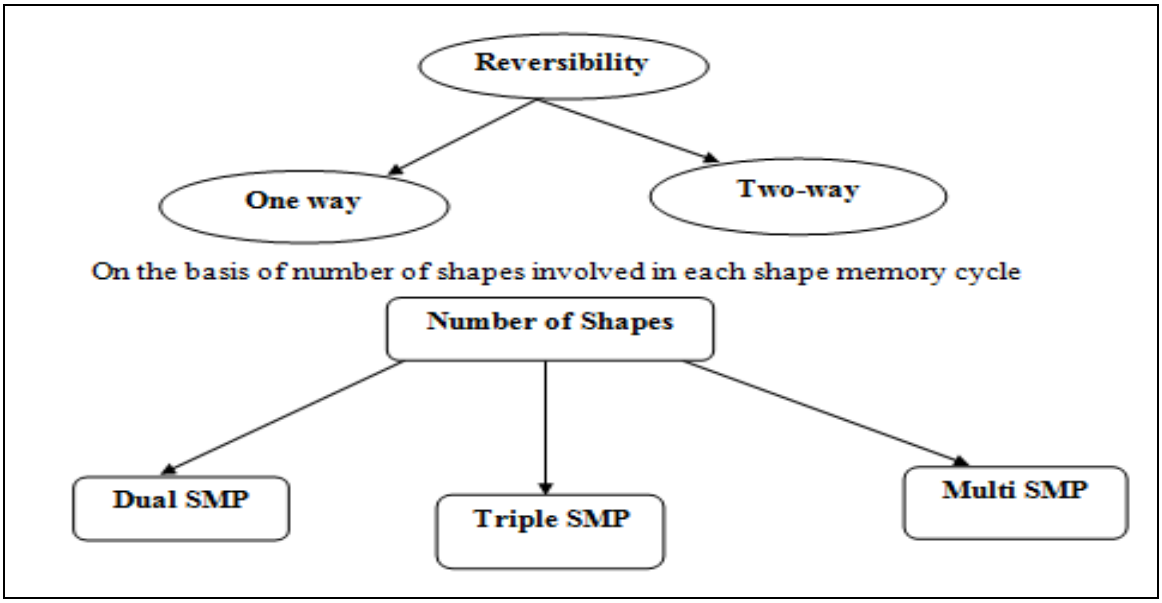
3. They have a Wide Range of Structural Designs: There is an abundance of methods for designing net points and switches for various sorts of SMPs. Additionally, specific polymers and foreign materials can be used to assemble one of a kind SMEs.

4. They Possess Tunable Properties: The two properties of SMPs can be engineered very easily and accurately tuned with the use of composites, blending, and synthesis methods.

5. They are Well-suited for Responses to Human senses/tissues and Biodegradability: SMPs are made from polymers, which are gentle materials that supply a considerable array of selections for making surprisingly biodegradable, biocompatible, and satisfied gadgets to interfere with our bodies, presenting unique possibilities for smart medical, organic and garment-integrated devices.

6. They can be very light and can Occupy a Massive Extent (Foam): these homes are extremely essential for applications, such as aerospace devices, air pressure items, and plane components.

Classification pf Shape Memory Polymer: The SMP are classified as follows: On the basis of the reversibility of shape memory effect [5-7].



On the Basis of Shape Memory Effect: One-way shape memory polymers: One-way implies that the shape healing is irreversible. At is, structure shifting for the duration of restoration can only proceed from a temporary to a permanent form and now not the reverse [13-15]. As more and extra programming protocols are devised and mentioned in the literature, one-way SMPs have been observed to be lots greater flexible, versatile, and applicable than the two-way SMPs because two-way SMEs are challenging to software or do not require programming. One-way SMEs have their unique shapes predefined in the manufacturing process, whilst brief shapes can be various based on exceptional shape-memory programming processes. There are quite a few types of current contrast strategies for characterizing one-way SMEs, such as cyclic tensile investigation [16], pressure recuperation system [24], bending checks [25, 26,] and shrinkage determination [27]. Considerable effort has been committed to finding out the elements that have an impact on the shape-memory performance of SMPs of one-way SMEs. Different affect elements have been studied using common

one-way SME structure(SMPU, SMPUU, and SMPU ionomers), such as the phase contents [18, 20, 22, 28], the molecular weight and crystallization of smooth segments [19, 28-29], ionic team contents [23], thermomechanical cyclic conditions *i.e.*, deformation and fixing pace [17] and maximum strain and processing conditions [30].

The section contents and the molecular weight of soft segments play a vital function on the shape-memory properties, specifically R_r, R_f and the structure healing stress. The SMPUs with 10 wt. % HSCs had no hard section domains, while these with 15 wt. % ≤ HSCs ≤ 40wt. % exhibited appropriate from recuperation (over 90%), whilst the structure restoration dramatically dropped as the HSCs elevated above 45 wt.% due to the hard segment domains changing from remote states into interconnected states.

The crystalline of the soft segment reduced and the inter-area spacing reduced, whilst the quantity and size of the hand-segment domains rose with growing HSCs. The recuperation stress can be adjusted by varying the HSCs. Specifically, the presence of inflexible chain extenders can end result in higher restoration stress [21]. Thus, achieving accurate one-way SMEs require a micro-domain morphological shape with difficult domains.

Additionally, the ample molecular weight of the soft segments, particularly in Tm-type SMPUs is a prerequisite for accomplishing correct shape–memory properties, and their superior molecular weight is typically 4000-6000 g/mol. Therefore, *via* various content materials of the smooth or challenging segments, the molecular weight of the gentle segments and the instruction process, we can modify the morphologies of SMPUs and for that reason, control the shape-memory properties.

Two Way Shape Memory Polymers: "Two-way" Workable that the Shape Change: is reversible; the preliminary and brief shapes can be reversed with the look and termination of the stimulus.

To these two-way SMPs can cut twin or even triple shape adjustments, two-way SMPs have obtained a proper sized interest in present-day years because of their capability to alternate shapes in response to the exterior stimuli to which they are exposed [31].

On the Basis of Number of Shapes Involved in Each Shape Memory Cycle: Dual shape memory polymers: The SMP is first heated to a deformation temperature (Td), which leads to the material softening (modulus drop). A deformation force is subsequently applied (*i.e.*, loading). The SMP is then chilled off under the heap.

Upon load removal (unloading), the deformed temporary shape is fixed, which marks the completion of the shape fixing. When the SMP in its temporary shape is re-heated to a recovery temperature (Tr) under a tranquil condition, the first (or perpetual) shape is recouped. In typical cases, both Td and Tr are above the reversible thermal transition temperature (glass transition temperature Tg or melting point Tm) of the SMP, which is thus called shape-memory transition temperature (T trans). In both **Fig. 2A** and **B**, a total of two shapes (one temporary shape and one permanent shape) are involved in each shape memory cycle [32-33].

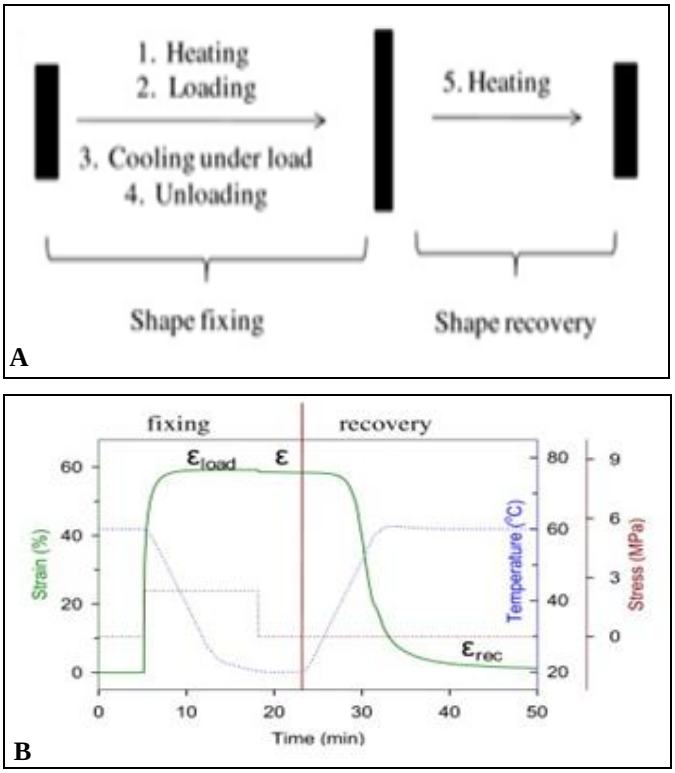
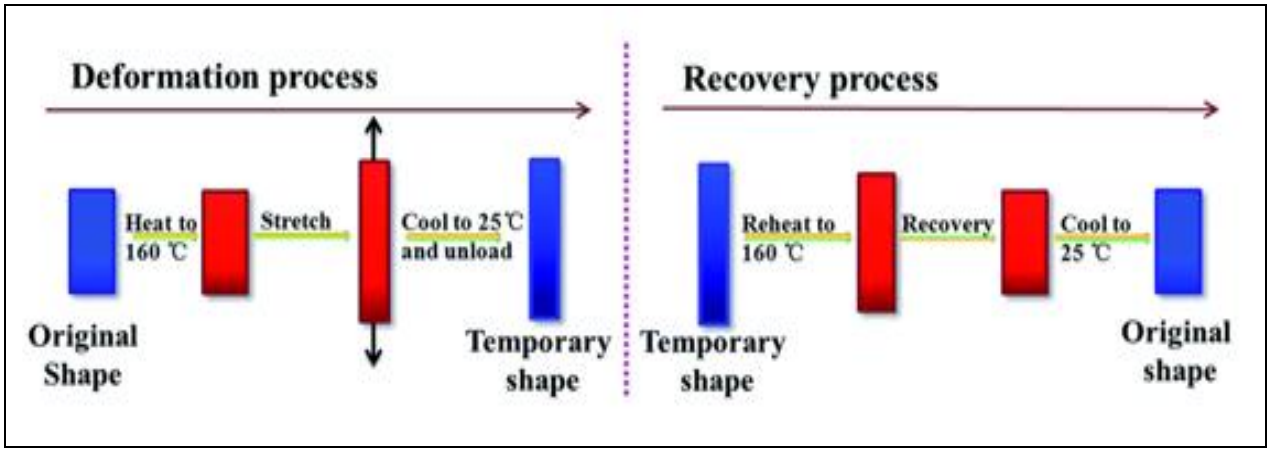


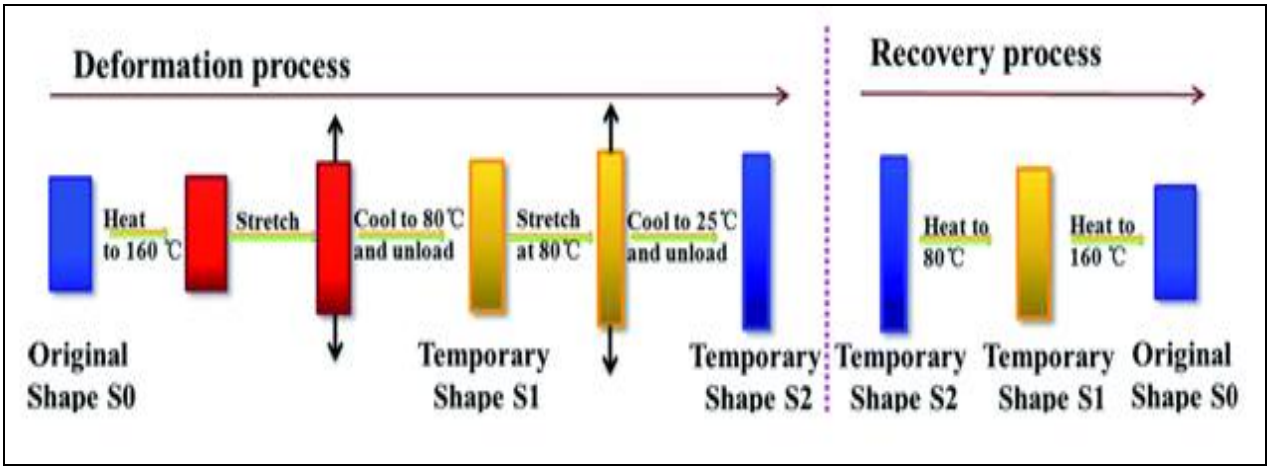
FIG. 2: CONVENTIONAL DUAL-SHAPE MEMORY CYCLE. (A) SCHEMATIC ILLUSTRATION AND (B) QUANTITATIVE TMA CYCLE

Triple Shape Memory Polymers: Triple SMPs are successful in fixing two brief shapes and recuperating sequentially from the first temporary structure (Shape a) to the 2-D brief form (Shape a),

and in the end to the everlasting structure (Shape a) upon a sure stimulus. Generally, a triple SMP is described to a multiphase polymer community that consists of at least two separated domains, which are associated with character transition temperatures. Thus far, triple SMEs have been carried out in chemically [34] or physically [35] crosslinked networks, LCEs [36,] and even in a gadget with a bi-layer structure.



A. DUAL-SHAPE MEMORY BEHAVIOUR



B. TRIPLE- SHAPE MEMORY BEHAVIOUR

FIG. 3: ILLUSTRATION OF THE SHAPE–MEMORY FUNCTIONALITY OF SMPS. (FOR ONE-WAY SME, ONCE THE STIMULUS IS TERMINATED, THE TEMPORARY SHAPE IS RETAINED, WHILE FOR TWO-WAY SME, THE TEMPORARY SHAPE CAN BE RECOVERED TO THE INITIAL SHAPE WHEN THE STIMULUS IS TERMINATED.)

Multiple SMP: Following the investigations of triple SMPs, used the extensive glass transition of Tg-type polymers to show a couple of SMEs, which can consider more than two brief shapes. They also systematically investigated the have an effect of thermo-mechanical stipulations on these a couple of SMEs of PFSA.

The results indicated that the programming and recuperation heating methods extensively affect the multiple shape memory houses when constructing multiple shape-memory cycles [37]. As two nano-fillers (Fe_3O_4 and CNT) can induce warmness at two distinctively exceptional radiofrequency tiers (296 kHz and 13.56 MHz), the multi-composite confirmed well-controlled a couple of shape recoveries [38].

The huge Tg vary attributed to the formation of semi-IPNs, which led to the partial miscibility of amorphous PMMA with PEG.

All of these publications on triple SMEs and more than one SMEs will not only meet the necessities of more and more complicated functions, however, will additionally further stimulate the development of shape memory features of SMPs.

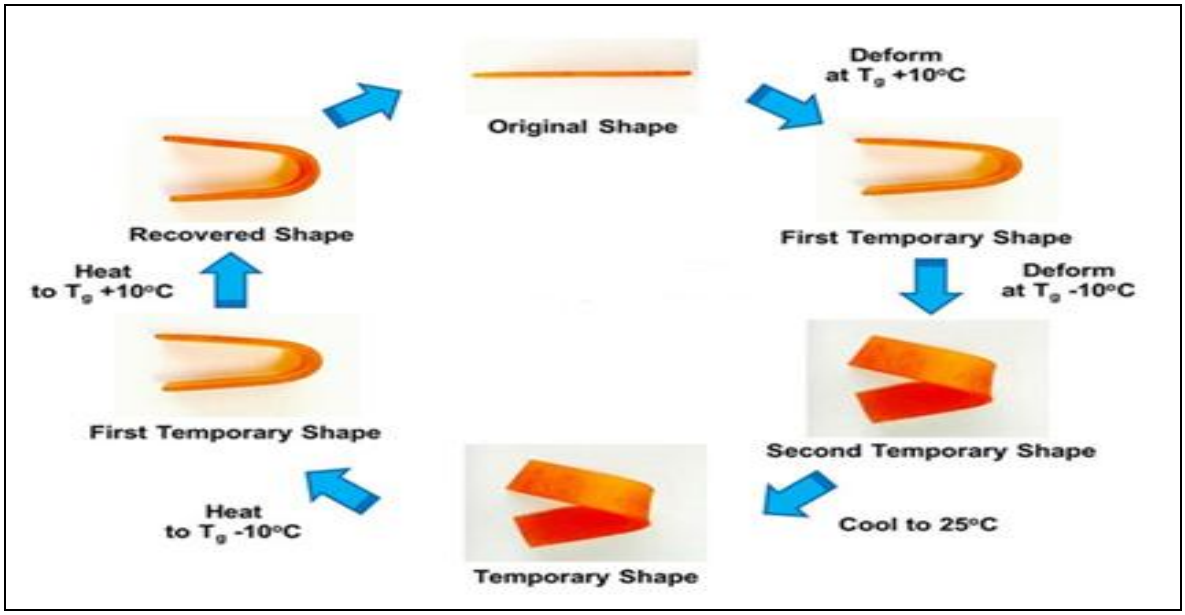


FIG. 4: MULTIPLE SHAPE MEMORY POLYMER

Classification and Methods of Shape Memory Hydrogel: [1]

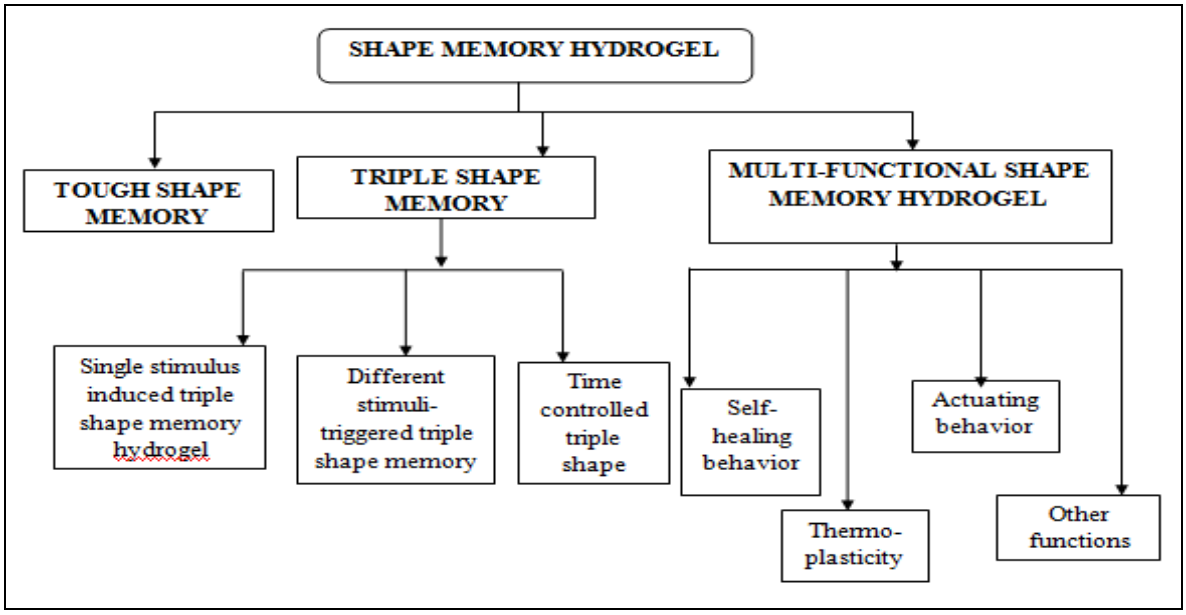


FIG. 5: CLASSIFICATION AND METHODS OF SHAPE MEMORY HYDROGEL

Tough Shape Memory Hydrogels: Because of the susceptible bonding strength of reversible interactions, SMHs based totally on only exchangeable bonds typically go through from negative mechanical properties. However, low power hydrogels would lose their tremendous practicable for use in load-bearing applications. Thus, there is a pressing need to improve the mechanical houses in a gorgeous way. Many techniques have been utilized to beautify the mechanical residences, which includes tensile strength, compressive strength, and toughness, for contributing to strong hydrogels, which can additionally be used for SMHs for instance, developing precise structures (double network, dual/triple crosslinks in a single network) or doping nanomaterial, *etc*. It is properly known that double community structures may want to enhance the mechanical homes effectively, in which one network acts as a sacrifice community for strength dissipation and the different serves as an elastic network to maintain everlasting shapes [39-43]. Similarly, SMHs normally have two unique crosslinks, either forming a physical community or chemical network, for fixing transient shapes and maintaining unique shapes. Thus, the reversible

switches used for the structure memory impact can additionally play a necessary section in strengthening the toughness of hydrogels. For example, a tough hybrid hydrogel having each physical and covalent crosslinks, and it can achieve shape reminiscence conduct *via* the switching of glassy nano-domains at a certain temperature [44].

By integrating a bodily crosslinked gelatin network and a chemically crosslinked PAAm network with graphene oxide (GO), Tong's team realized NIR-triggered form memory performance, in which both the gelatin networks and the GO bridging take part in dissipating deformation energy [45] According to a similar mechanism of energy dissipation, the construction of dual/triple crosslinks in a single network is another wonderful strategy to render SMHs with suitable mechanical properties.

Triple- / Multi-shape Memory Hydrogels: Previous SMHs, processing a dual shape memory effect, can only take into account one easy brief structure in every memory cycle. Since the wide variety of transient shapes that can be fixed will affect the durable purposes of SMHs, more and extra interest has paid to the fabrication of triple / multi-shape reminiscence hydrogels.

Nowadays, there are three essential approaches to endow SMHs with triple-/multi-shape memory effects, which are summarized in the following.

Single Stimulus-induced Triple-shape Memory Effect: Responses of SHMs to a single stimulus may be realized when exclusive brief crosslinks feature various stages of sensitivity to one stimulus, ensuring in triple/multi- shape-memory performance. An early consultant instance is with the aid of in which the first SMH with a triple structure reminiscence impact was once exposed **Fig. 6**. The association and dissociation of each dipole–dipole interactions between nitrile companies (CN–CN) and Zn–CN linkage can be adjusted with the aid of the attention of zinc ions, leading to a triple shape memory behavior. As one of the most usually used stimuli, warmth can be employed to plan SMHs with a triple/multi-shape memory effect due to the extraordinary forces generated from distinct interactions. Hydrogels reported by are composed of two specific kinds of hydrophobic crystallizable switching temperatures. During the programming system of altering the temperature, two wonderful temporary shapes can be memorized and recovered *via* crystallization and amorphization of hydrophobic side chains [46].

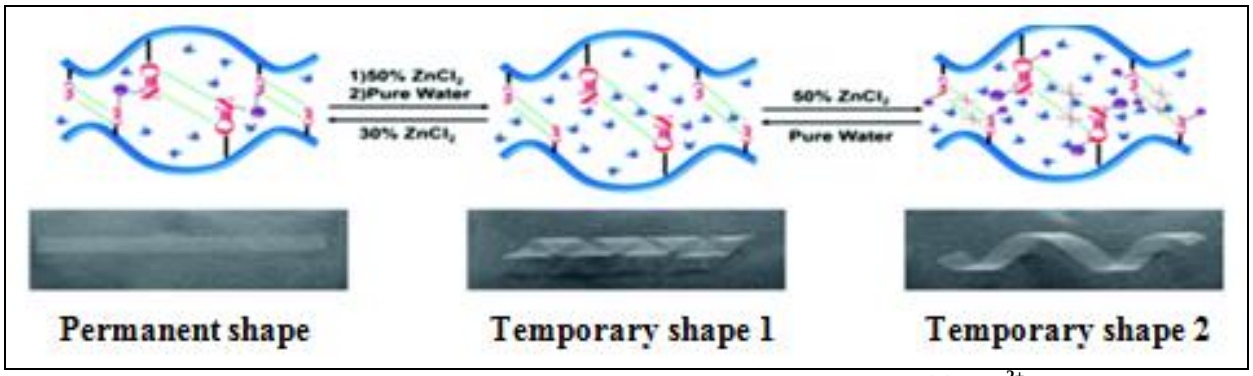


FIG. 6: SINGLE- STIMULI- INDUCED TRIPLE- SHAPE MEMORY EFFECT (1) ZN^{2+} TRIGGERED TRIPLE MEMORY EFFECT BY MANAGING THE INTERACTIONS OF CN-CN AND ZN-CN. (2) HEAT-TRIGGERED TRIPLE SHAPE MEMORY EFFECT AT DIFFERENT TEMPERATURES BECAUSE OF TWO OF SEMI CRYSTALLINE SIDE CHAINS

Different Stimuli-triggered Triple-/Multi-Shape Memory Effects: As special stimuli can be brought step through step, combining non-interfering interactions in one material presents an environment-friendly way to create triple/multi-shape memory materials. Typical examples are the stretchable supermolecular hydrogels with a triple-shape memory impact such as those fabricated **Fig. 7-I**. By sequentially introducing two independent interactions, together with alginate-Ca^{2+} chelation and phenylboronic acid (PBA)- diol ester bonds, into a double- network system, an impressive triple-shape memory overall performance has been successfully finished at both the macro-scale [47]. In addition, utilizing one interplay for the shape memory impact and every other interaction for self-healing, form reminiscence after the self-healing process or self healing technique for the duration of

shape memory performance can also be realized. Similarly, Schiff base bonds and metal coordination interactions can additionally be built-in for realizing the triple structure memory properties [48]. The reversible Schiff base bonds between the amino agencies of chitosan and aldehyde organizations of oxidized dextran may want to be applied to memorize temporary shapes. Meanwhile, number of metallic cations can additionally chelate with chitosan for fixing different brief shapes. As proven in **Fig. 7-II**, a light-, pH-, and thermo-responsive hydrogel with a triple-shape memory impact was organized with the aid of introducing dansyl-aggregations and host-guest interactions on the groundwork of azo benzene (Azo) with cyclo-dextrin (β-CD). Since the aggregation of dansyl corporations at an excessive pH charge and the supramolecular inclusion complexes of β-CD-Azo underneath seen the light can act as reversible switches, a triple-shape reminiscence impact can be realized in response to light and pH sequentially.

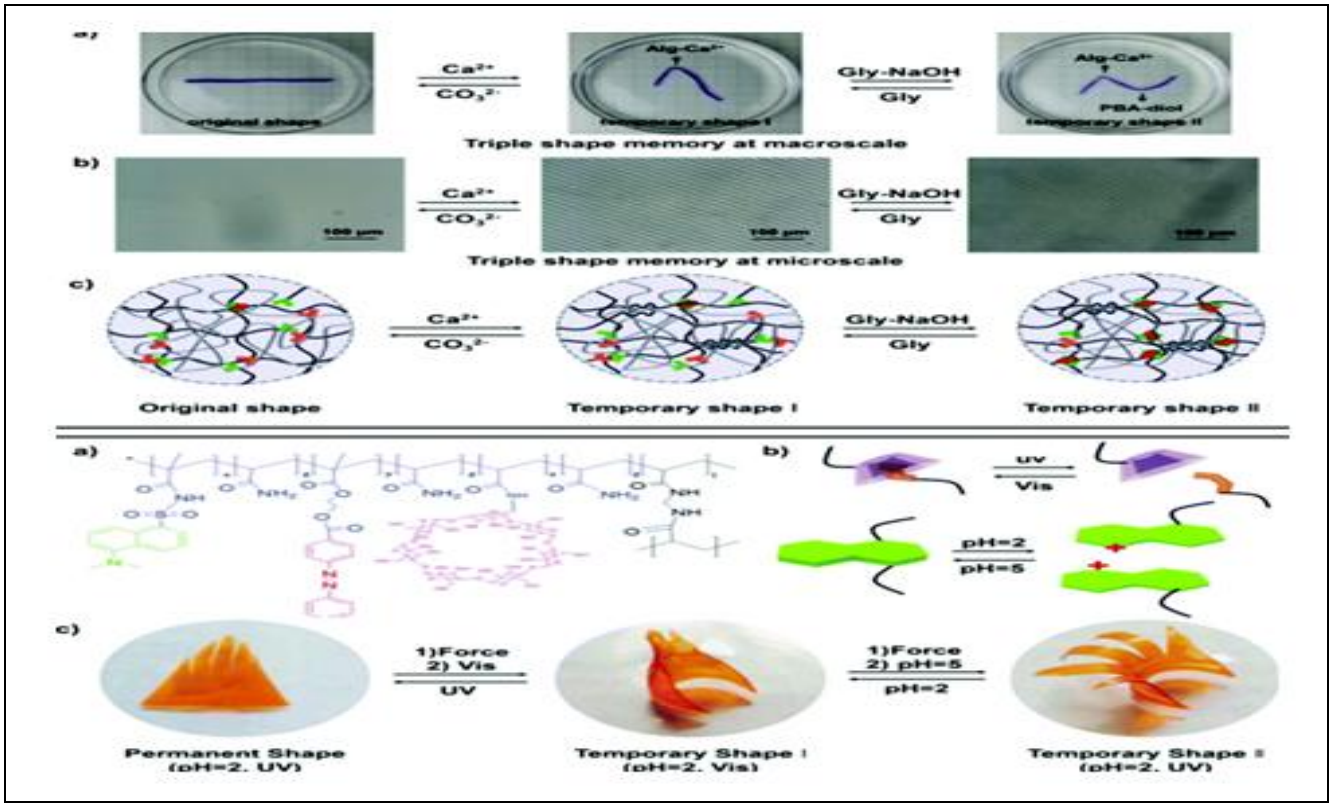


FIG. 7: DIFFERENT STIMULI- TRIGGERED TRIPLE-SHAPE MEMORY EFFECTS.(I) TRIPLE- SHAPE MEMORY EFFECTS AT BOTH THE MACRO-SCALE AND MICRO-SCALE HAVE BEEN ACHIEVED BY PROGRAMMABLE INTRODUCTION OF ALG-CA^{2+} CHELATION AND PBA-DIOL ESTER BONDS. (II) TRIPLE SHAPE MEMORY EFFECT REALIZED BY ADJUSTING PH AND UV/VIS STIMULI

Time-controlled Multi-shape Memory Effects: Another easy method to recognize a multi-shape reminiscence effect is through adjusting the processing time. By altering the time for memorizing shapes, distinctive temporary shapes can be fixed. As an instance for a time-controlled multi-shape memory, a PAA–Go–Fe^{3+} hydrogel with a twin physically crosslinked structure was once synthesized [49] **Fig. 8I**. By adjusting the immersing time in $FeCl_3$/HCl and pure water, for one of a kind transient shapes have been constant due to the kA Ac) (mono-, bi- and tridentate). Furthermore, with the aid of changing the soaking time in HCl, the form memorized hydrogel recovered sequentially to its unique shape. Another normal instance was by chen's group [50]. Reversible physical interactions, together with chitosan micro-crystals and chain-entanglement crosslinks, can be performed virtually by soaking chitosan in NaOH and NaCl solutions, respectively.

Due to the diffusion transition mechanism, the stimulating chemical (NaOH solution or NaCl solution) diffuses from outdoor of the hydrogel to the inside for fixing transient shapes step *via* step, enforcing a multi-shape memory impact **Fig. 8II**.

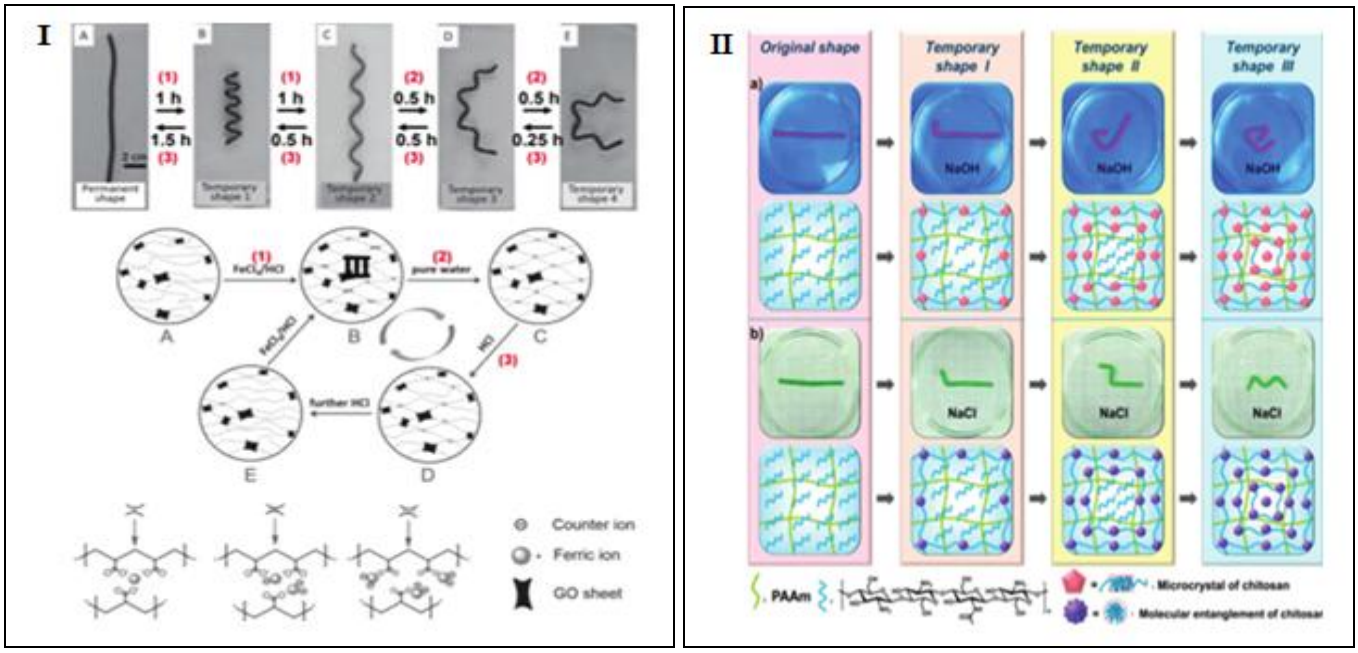


FIG. 8: TIME-CONTROLLED MULTI-SHAPE MEMORY EFFECT. (I) ADJUSTING THE TYPES OF FORMATION BETWEEN FE^{3+} AND AAC (MONO-, BI- AND TRIDENTATE) BY CHANGING THE IMMERSING TIME IN A FECL3/HCL OR HCl SOLUTION FOR MULTI-SHAPE MEMORY (II) DIFFERENT DEGREES OF FORMATION OF MICRO CRYSTALS OR MOLECULAR ENTANGLEMENT OF CHITOSAN FOR MULTI-SHAPE MEMORY

Multifunctional Shape Memory Hydrogels: Since a single characteristic can often not meet all requirements for practical applications, it is pretty promising to develop SMHs with multi-functionalities, such as self-healing, self-adhesion, thermo-plasticity as properly as antibacterial and anti-inflammatory functions.

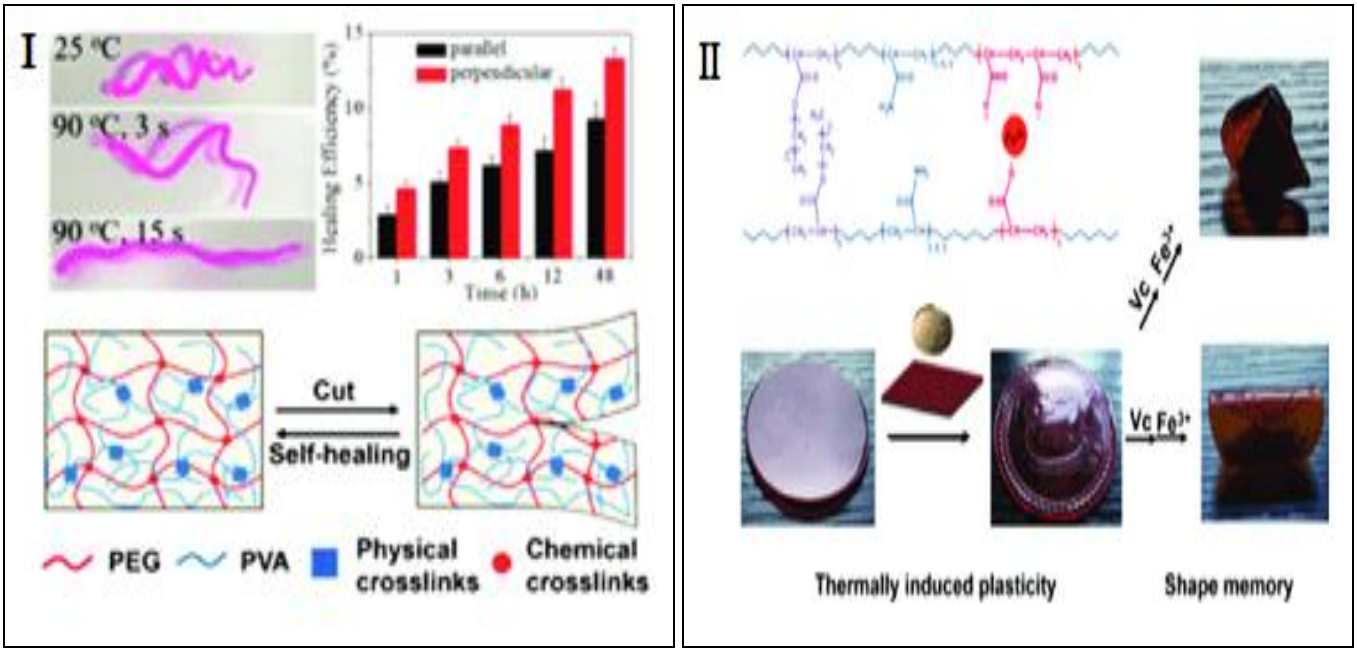


FIG. 9: SMHS WITH OTHER FUNCTIONALITIES. (I) SMHS WITH SELF-HEALING BEHAVIOUR (II) SMHS WITH THERMO-PLASTICITY

SMHs with Self-healing Behaviour: Since the service lifestyles of materials is an vital index for materials, self-healing materials can fix their functionalities after damage, which is additionally of terrific importance for SMHs. Experts have done a full-size quantity of research to endow SMHs with a self-healing performance. First built a supra-molecular hydrogel with each self-healing and form

reminiscence properties [51] which was based on dynamic PBA-diol ester bonds and the formation of an alginate- Ca^{2+} complex, respectively. A double community hydrogel composed of a poly(ethylene glycol) (PEG) community and a poly(vinyl alcohol) (PVA) network [52]. The former chemically crosslinked network was used to maintain the everlasting shape, and the latter one, which was physically crosslinked, can be applied for both structure memory, and self-healing **Fig. 9I** as the two bonding motifs do not intrude with every other.

SMHs with Thermo-plasticity: Among all the functions, thermo-plasticity is a specifically appealing one. Once thermo-plasticity is introduced into SMHs, the everlasting shape can be changed, which means SMHs can possess a variety of permanent shapes, main to more brief shapes. A hydrophobic poly-ampholyte SMH reported a special thermo-plasticity, which can change its everlasting form upon heating [53]. Through either ligand-ion bindings or salt-dependent hydrophobic association, transient shapes can be memorized. Recently a novel dual move linked single-network hydrogel has been fabricated *via* co polymerization of acrylamide (AAm) and AAc with n-octadecyl acrylate [54]. Without any chemical cross-linkers, the hydrogel exhibited a fascinating thermo-plasticity. In addition, alternative use of vitamin A and Fe^{3+} triggered the structure reminiscence and the shape recuperation system **Fig. 9II**.

SMHs with Actuating Behaviour: Up to now, the transient shapes of now a day present SMHs are usually created by guide deformation. However, this simple approach will be confirmed when it comes to the education of complicated transient shapes, which is hard to manipulate by means of hands. Inspired by means of the sketch thinking of hydrogel actuators, SMHs with actuating behavior have been reported. For example, a bi-layer form reminiscence hydrogel has been fabricated inside a thermo-responsive actuating layer and a pH-responsive memorizing layer [55]. in the system, one layer made of PNIPAM hydrogel shrank at greater temperature, main to the self- deformation of the total hydrogel. After that, the different layers containing chitosan could form micro-crystals for stabilizing transient shapes when soaking in base solution. By the use of photo-masks, greater complicated brief shapes may want to be done due to nearby deformation. In addition, every other anisotropic hydrogel with built-in-self-deformation and controllable structure memory effect has also been mentioned. Through establishing an anisotropic PAAc–PAAm structure, they got hydrogel deformed into sure shapes in response to pH stimulus, which can be constant by the coordination between carboxylic organizations and Fe^{3+}. Moreover, the form memory ratio and form healing ratio should be adjusted with the attention of the corresponding ions, such as Fe^{3+} and H^{+} [56].

SMHs with Other Functions: In order to adapt to complex situations, SMHs are now designed to have extra and more functions. A novel supra-molecular hydrogel possessing a self-healing function, a form memory and an adhesion property has been reported [57]. Since Ca^{2+} can chelate with alginate for retaining everlasting shapes, the affiliation and dissociation of dynamic phenyl boronic acid (PBA) catechol interactions should be utilized for a structured memory and self-healing performance. In addition, the catechol moieties endow the hydrogel with perfect adhesive properties. Besides structure memory, antibacterial and anti-inflammatory features have been integrated to fabricate multi-walled hydrogel tubes for a conceivable tissue engineering scaffold [58].

Mechanism of Shape Memory Hydrogel: The shape reminiscence mechanism in polymers is primarily based on a twin material. On the other hand, an elastic polymer community with net factors is required to outline the everlasting shape. These factors normally are chemical cross-linking or bodily internet points such as crystalline domains or complexes. In addition, a programming technique consisting of the elastic deformation of the sample takes locations, and additional molecular switches furnish brief crosslink's to reversibly restorative the brief form of the material [3].

As a result of the application of an external stimulus, typically temperature, molecular switches are disturbed, and the polymer chains acquire their initial mobility that leads to a macroscopic movement resulting in the initial form [59]. In the case of thermoresponsive SME polymeric segments with specific thermal transitions like glass transition, melting, or liquid- crystalline section

transition, these transitions can act as molecular switches when the temperature is varied. In order to extend the possibility of SMPs as biocompatibility, mechanical properties and biodegradability have promoted the improvement of the hydrogels with SME. Like thermally incited SMPs, supra molecular structure memory hydrogels existing cross-linking that characterize the system, this is the perpetual shape and boosts responsive switches, comprising of reversible co-operations to restore the transient structure **Fig. 10**

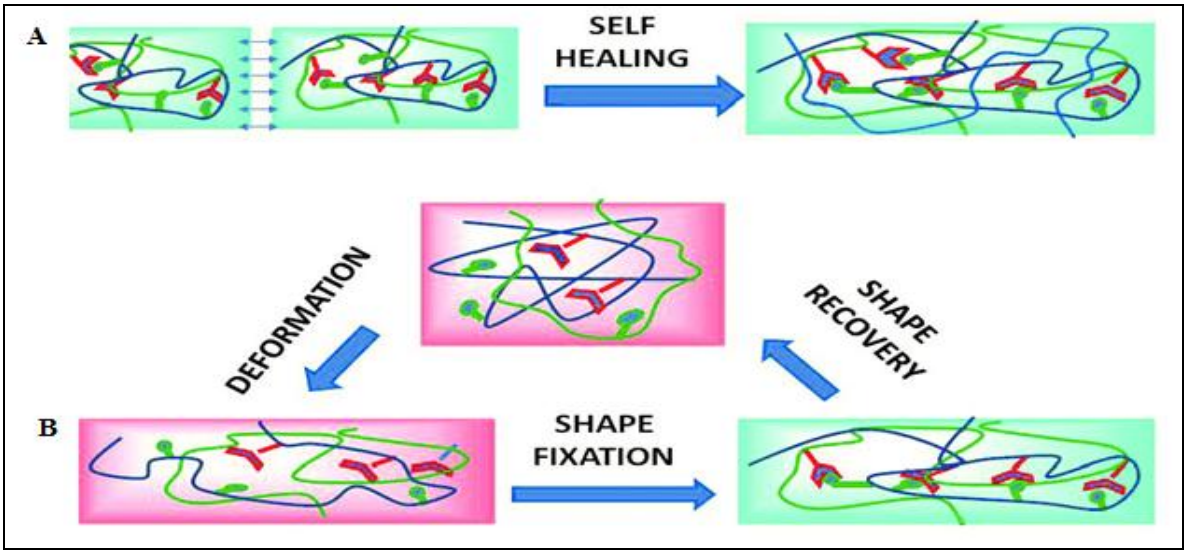


FIG. 10: SCHEMATIC REPRESENTATION OF (A) SELF-HEALING AND (B) SHAPE MEMORY EFFECTS FOR HYDROGELS

Thus, form fixation and healing demand interactions effortless to be damaged and formed, whilst adequate mechanical properties of the fabric are intently related to the awareness and electricity of the possible types of cross-linkings. The temporary structure can be created by way of folding, elongation or compression. The excessive water uptake in hydrogel gives an upward squeeze to structure reminiscence substance able to undergoing massive deformation between the temporary and permanent states. Molecular switches, in the case of hydrogels, commonly are not section of the main chain forming the polymer network.

Typically, are pending moieties such as brief crystallizable side chains [60] unique agencies for host-guest interactions, [61] complex-forming groups [62] or organizations able to shape dynamic bonds [63; s]imilar to self-healing process, polymeric segments reorganize and water flows through the polymer network by using diffusion, and the kinetics of these systems in different states of deformation of the system will administer the worldwide pace of the SME. The simplest SME is also referred as dual shape reminiscence impact and corresponds to the case in which only one reversible interplay and consequently one temporary shape is constant in every form reminiscence cycle. Accordingly, triple form memory impact is corresponding to two reversible and impartial interactions happening in the equal hydrogel network [64-67].

Currently, regardless of the massive investigation effort made in the final years, nevertheless there is a decreased bibliography about triple and multi-shape memory hydrogel. Although non-covalent interactions are weaker than covalent interactions, their importance in polymer science has been shown in this context, supra molecular shape memory hydrogels based on non-covalent interactions present an arisen importance.

The incorporation of non-covalent interactions into hydrogel network ought to adjust the houses of the shaped hydrogel. These editions multiplied the feasible purposes not solely shape memory effect or a carefully associated self- healable ability, however additionally for the development of high tough hydrogels. In **Fig. 11**, the most common interactions used in structure reminiscence hydrogels are described, such as hydrogen bonds, metal-ligand interactions, and ion-ion interactions.

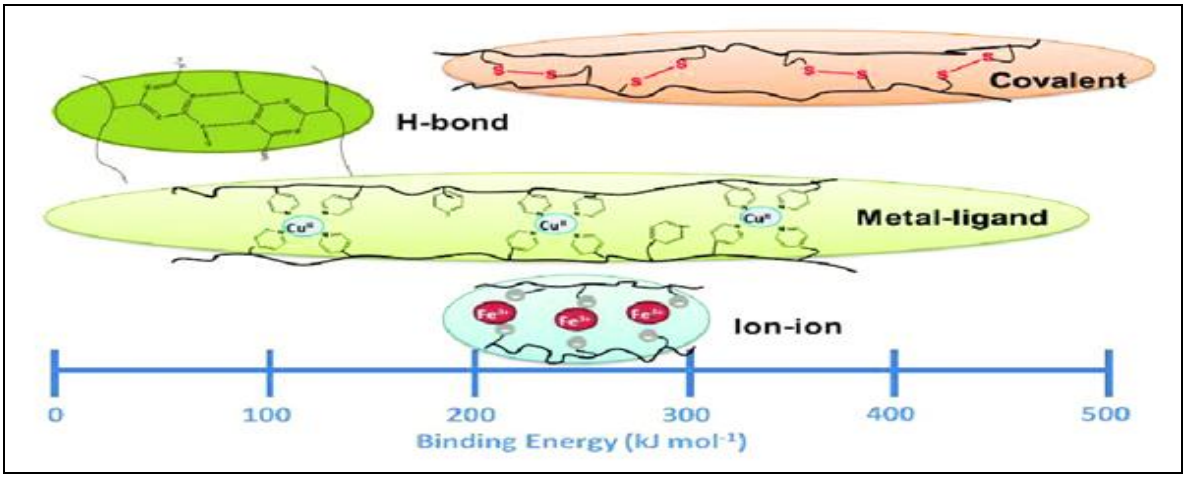


FIG. 11: REVERSIBLE INTERACTIONS FOR SHAPE MEMORY HYDROGELS CONSTRUCTION

Characterization:

Fourier Transform Infrared Spectroscopy: Fourier Radically Change Infrared Spectroscopy: Was used to decide the chemical structures of the organized Yb(TTA)2AAPhen and Nd (TTA)2AAPhen as well as the raw materials of AA, HTTA, and Phen.

The samples were ground with potassium bromide (KBr) collectively and pressed into a checking out tablet [68].

Reflectance and Transmittance Measurements: The reflectance spectra of Yb(TTA)2AAPhen and Nd(TTA)2AAPhen as nicely as the transmittance spectra of the copolymers had been measured with the aid of a UV–vis–NIR spectrophotometer (UV-3101PC, Shimadzu Corp., Kyoto, Japan), the usage of $BaSO_4$ and air as the references, respectively.

Thermo-gravimetric Analysis: In an airflow, the thermal steadiness of Yb(TTA)2AAPhen and Nd(TTA)2AAPhen as well as the copolymers hadbeen evaluated using thermo-gravimetric analysis (TGA, Netzsch SAT 449 ºC, Selb, Germany) scanning from room temperature to 800°C at the heating rate of 10 °C min^{-1}.

Thermal Properties Analysis: Differential scanning calorimetry (DSC, 204 Phoenix, Netzsch, Selb, Germany) was once used to determine the thermal residences of the copolymers. The scanning temperature elevated from room temperature to a 150 °C, and lowered to-75 °C earlier than increasing to a 150 °C again. The heating and cooling were performed below a nitrogen atmosphere at a rate of 10 °C min^{-1}.

X-ray Diffraction: The structure characterization of Yb (TTA) 2AAPhen and Nd (TTA) 2AAPhenpowders as properly as the copolymers had been carried out the use of a Smartlab (Rigaku, Tokyo, Japan) thin-film diffractometer using Cu Kα radiation (λ = 0.15046 nm), with the 2Θ attitude from 5°to 50°at the scanning rate of 10° min^{-1}.

Microscopy: The morphology of Yb (TTA) 2AAPhen and Nd (TTA) 2AAPhen powders as nicely as the copolymers were examined with the usage of a scanning electron microscope (SEM, JSM-6510, JOEL, Tokyo, Japan) outfitted with a NORAN System 7 EDX detector (Thermo Fisher Scientific, Pittsburgh, PA, USA).

Photo-thermal Effect: The NIR lights of 980 and 808 nm had been generated respectively by a laserdiode driver (KS3-11312-912, BWT Co., Beijing, China) and a laser driver (FC-808-10W, Xinchanye Co., Changchun, China). The sample temperatures had been measured using a hand-held infrared camera (Xintest Company, Dongguan, China). The energy density used to be determined the usage of an optical power/energy meter (Model 1918-R, Irvine, CA, USA) equipped with a thermopile detector (Model 818P-020-12, Newport).

Mechanical Properties: A flexural take a look at the instrument (MZ-2000c, Mingzhu Testing Machinery Co., Jiangdu, China) used to be used to operate the flexural test by using a three-point bending setup with a 30-mm span. The applied pressure rate used to be 1.5mms^{-1}.

The sample dimension used to be L × W × D = 60 mm × 15 mm × 0.9 mm. The flexural yield electricity and flexural modulus have been bought based on the yielding factor.

Shape Memory Effect: The shape – memory effect used to be determined on the basis of attitude variation; take a look at specimens had been placed onto a hot plate at 150 °C after 5min, the specimens had been completely folded to 90° in opposition to a perpendicular glass plate tightly and stored for some other 5 min before gradual cooling to room temperature. Upon the irradiation of NIR mild to the bended corner, the instantaneous angles, Ɵ between the real-time region and the primary location of the moving part have been characterized with the error of ± 2°. The form healing ratio, Rr used to be determined as Rr = Ɵ/90° × 100%.

Applications of Shape Memory Hydrogel: In general, SMPs are mainly used as sealants and self-healing materials, vibration control systems for structural health monitors.

Sealants and Self-healing Materials: An SMP-based smart sealant for compression-sealed joints in concrete pavement structures [69]. They also developed SMP-based syntactic foam that is cored with sandwich structures for the purpose of over and over self-healing, the have an effect on image [70].

Therefore, using SMPs as sealants (such as SMP-based, asphalt-based liquid, and two-way shape-changing polymer sealants) has grown to be a necessary application path in civil engineering. In addition to sealant applications, the self-healing abilities of SMPs have been additionally used to shape SMP-based composite structures, any other important software in civil engineering [71].

Vibration Control Applications: SMP-based structural factors (beams, rods, plates, composites, *etc*.) permit for the tuning of a range of frequency bandwidths and damping properties for vibration manipulate functions [72, 73].

The fabrication procedure and dynamic vibration is checking out of an electrically activated SMP [74]. They confirmed how SMP beams might want to acquire variable stiffness and damping with a reasonable.

The mechanical and damping residences of SMPs exhibit that SMPs can be used as damping materials, opening the door to vibration control applications in earthquake engineering [75].

Biomedical Applications:

Drug Delivery: To supply drugs, the porous shape of hydrogels can provide a matrix for drug loading and protect pills from opposed surroundings at an identical time. Moreover, this porosity can be managed by means of various moves linking density of the gel matrix. The launch rate, some other necessary parameter for drug carriers, primarily relies upon the diffusion coefficient of this molecule through the gel community and can additionally be tuned in accordance with specific requirements. Biocompatibility and bio-degradability can be acquired through designing sure chemical and physical structures for hydro-gels. All of these residences hydrogel tremendous *via*ble to be used for drug delivery [76, 77].

Hydrogels for Three-Dimensional Cell Culture: Hydrogels, with excessive water content as nicely as tissue-like mechanical properties, have been verified to be successful of combining with cells to engineer a range of tissues in both *in-vitro* and *in-vivo* [78, 79]. A fundamental requirement for the construction of three- dimensional regenerative tissue in adequate quantities is an artificially created surroundings that enables biological cells to grow or interact with their environment in all three dimensions.

Medicine to Soft Robotics: The ability of hydrogels to comprise bioactive molecules for managed delivery [80, 81], being of activity particularly for sensitive factors that demand aqueous environment, can be accelerated to a stimuli-sensitive anchoring of drug-loaded devices in body cavities, for example, after oral administration or after minimally invasive implantation in the abdomen, urogenital or vaginal tract. Spatially directed movements of SMHs at the macro-scale or micro-scale may additionally be used to seal containers after filling with sensitive compounds, that is, the use of the form healing *ex-vivo* before implantation [82]. In particular, hydrogels should be used in soft robotics, which describes engineered mobile machines created by means of smooth materials. In contrast to traditional robots

basing on tough elements (*e.g.*, steel) and enabling directed movements *via* electric powered motors, soft robots are suitable for functions in herbal environments while preventing mechanical damage of their environment. In the case of gentle robots from SMHs, the activation manner can be realized by way of the SME offering purposes as pneumatic actuators, grippers, and microswimmers.

Especially soft robots composed of hydrogels signify an upcoming type as their actuation is not confirmed to the stimuli potentially relevant to non-swollen system. Here, actuations can be realized that require diffusion process, that is, ion-or signaling molecule- mediated shape switches as, proven for SMHs being touchy to ions, salts, glucose, modifications in pH value or redox potential [83] in order to allow SMHs to enter into their fields, switching kinetics, created switching forces, and reversibility of switching are elements to be viewed and in addition explored [84]. Switching mechanisms realized for hydrophobic SMPs or plan principles-based totally on multiple materials as in an SMP-coated synthetic muscle 85 may also supply idea on this path.

Future Outlook: The research regarding shape-memory polymers continues its fast growth, with current efforts divided between enabling applied sciences (materials, processes, and techniques) and utility identification and prototyping. On the enabling front, there are growing quantities of reviews discovering novel healing set off mechanism different than exterior heating. Examples consist of photo, magnetic and humidity. In another example, one of us has conceived a mechanically-activated shape-memory device; the place the exothermic warmness from the crystallization of a super-cooled liquid held inside a SMP container is transferred to the SMP to stimulate structure recovery. While some of these non-traditional shape- memory techniques are definitely nonetheless thermally triggered, for example through warmness from irradiation or magnetic fields, some are caused by means of a trade in fabric properties caused either by means of the photo-isomerization of the constituent molecules or a plasticizing effect after the material is swollen in water or different media. Aside from alternative triggering mechanisms, there is additionally an emerging pastime in shape-memory polymers capable of memorizing two or more transient shapes. At exceptional temperature stages, this variety of SMP can recover in a tandem manner from one shape to any other and eventually to its unique shape. This type of SMP, referred to as ‘‘tandem shape-memory polymer’’ or ‘‘polymeric triple-shape materials are anticipated to be successful of offering greater complex actuation activities and workable for utility in biomedical devices or deployable structures.

CONCLUSION: This review summarizes the current tendencies of the shape memory polymers in our group and other main research groups, along with their applications in the medical field. In this paper, a new classification scheme has been adopted primarily based on the mechanisms of form fixing and structure recovery, and this is further used to organize the review. Also, a new method closer to the quantification of the shape-memory capacity was once proposed, and applications and future outlook of SMPs were described.

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