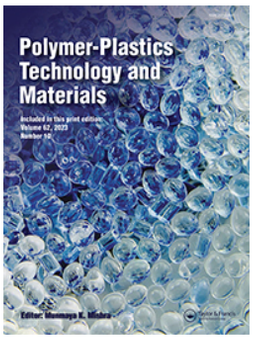
Construction, characterization, properties and multifunctional applications of stimuli-responsive shape memory polymeric nanoarchitectures



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Construction, characterization, properties and multifunctional applications of stimuli-responsive shape memory polymeric nanoarchitectures: a review

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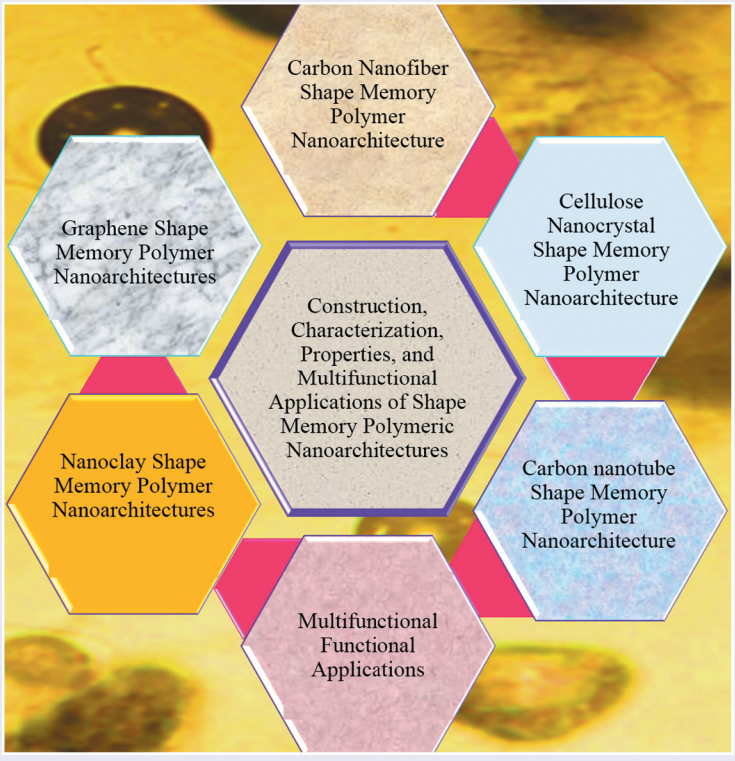
ABSTRACT

Due to the advent of nanotechnology, deficiencies and limitations inherent in stimuli-responsive shape memory polymeric matrices (SMP), have been effectively mitigated, through the inclusion of a versatile range of organic or inorganic nanoparticulates within the confines of SMP matrice/s. This phenomenon has resulted in the emergence of shape-memory polymeric nanoarchitectures (SMPNs) possessing enhanced and outstanding properties, when compared with the pristine SMP, and this has subsequently enlarged their scope of applications (civil engineering, biomedical gadgets, aerospace, bionics engineering, energy, electronic engineering, household products, and textile engineering). Furthermore, SMPNs enhances athermal stimuli-activities including electroactivity, magneto-activity, water-activity, and photo-activity, as well as shape memory effect (SME) including multiple-shape memory effect (MSME), spatial shape memory effect (SSME), as well as dual-route shape memory effect (DRSME). This elucidation is essential and imperative at this time to enlighten the polymeric universe on new advancements in fabrication, features and applications of stimuli responsive SMPNs. Therefore, this paper, presents, very recently emerging advancements, in construction, characterization, properties and multifunctional applications of stimuli-responsive SMPNs with special emphasis on carbon nanotubes (CNT), carbon nanofibers (CNF), cellulose nanocrystals, and nanoclay reinforced SMPNs.



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1. Introduction

Stimuli-responsive polymeric matrices remarkably vary their attributes including phase separation, shape, mechanical behaviors, surface, permeability, optically inclined properties, as well as electrical attributes when subjected to small changes in environmental parameters relative to electric field, temperature, pH, glucose, light, electrical field, magnetic field, sonic field, ions, solvent, as well as enzymes as depicted in Figure 1.[1–7]

Shape memory polymers (SMPs) are a class of stimuli responsive polymeric matrices, capable of recovering their original (or permanent) shape when subjected to external stimuli.[8–14] SMPs are capable of versatile utilization in varying areas including biomedical gadgets, textiles, aerospace, energy, electronic engineering, bionics engineering, civil engineering, as well as household items. Table 1 summarizes prevalent applications of SMPs in biomedicals, while Table 2 presents SMP applications in functional textiles.

SMPs are composed of a permanently fixed and temporarily shaped configurations resulting from a synergy of molecular chemistry as well as a programming outlay.[42] The necessary chemical architecture entails a net-point as well as a molecular switch sensitive to stimulus. Similarly, SMPs may be perceived as copolymeric matrices composed of a hard and soft components functioning as a fixed and reversible phase, respectively. The fixed phase hinders polymeric chains flowing on stress application while the reversible phase exhibits deformation and molecular switching which freezes the temporary shape on stimulation while returning to the original configuration on stimulation switching. The mechanisms of the molecular disposition of SMPs architecture is elucidated in Scheme 1, and capable of being applied to differing forms of SMPs.[42]

Here, SMPs are composed of both net-points as well as molecular switches. This fixed phase or net-points are garnered through inclusion of inter-locked supramolecularly inclined complexes, crystalline segments, chemical cross-linkages, interpenetrating networks, and chain entanglements. The switch portions take care of shape fixity as well as recovery upon application of an external stimulus. Thus, crystalline, liquid crystalline, amorphous segments, supramolecular substrates, light-reversible synergies, along with percolating cellulose-whiskers have been incorporated as switching entities in SMPs.[42] The phase of reversibility fixes the temporary configuration via crystallization, glass transition, isotropic transition, supramolecular interactions, reversible covalence or non-covalent bonding.[42]

On the other hand, SMPs exhibit intrinsically inferior mechanical strength, low shape recovery stress, poor stiffness due to low rubbery moduli, elongated time of recovery, poor responsivity as a result of inferior thermal conductivity, along with electromagnetic unresponsivity to stimuli, ascribed to electromagnetic insulation of most polymeric matrices, which have minimized the scope of SMPs applications. Hence, in order to mitigate these challenges, smallish levels of nanoreinforcement/nanomaterials/nanofillers or nanoparticulates have been embedded within SMPs matrices resulting in enhancements in mechanical behavior and shape recovery stress of SMPs, and so on, due to the formation of shape memory polymeric nanoarchitectures (SMPNs). Additionally, due to nanoreinforcement effect, SMPNs can improve athermal stimuli-active

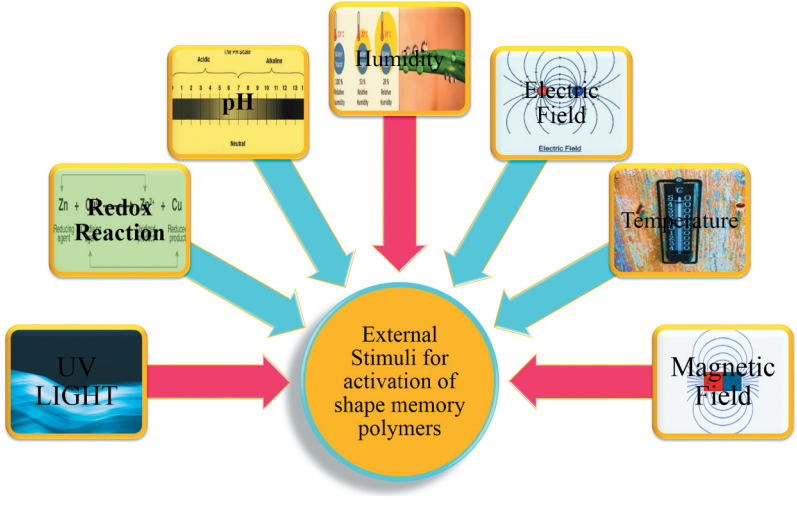


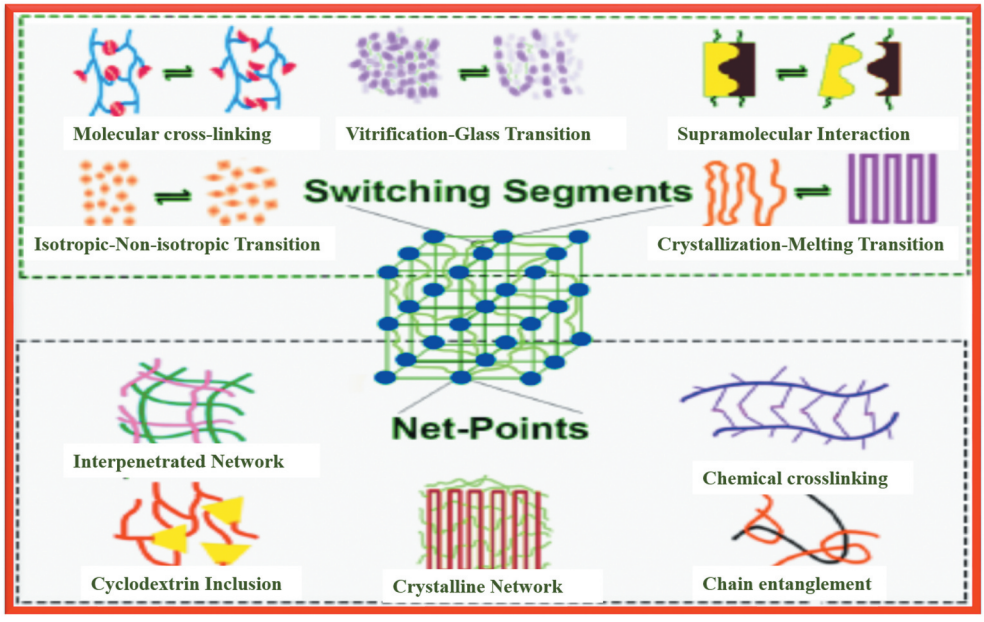
Figure 1. External stimuli affecting SMPs.

Table 1. Biomedical applications of shape memory polymer nanoarchitectures.[8–30]

Biomedical applications	Ref
Wound dressing	8
Surgery within living cells	9
Vascular stents	10
Esophageal stenosis treatment	11
Skin-care products	12
Shape memory neuronal probe	13
Post-surgical treatment of mitral insufficiency	14
Reconstruction of pharyngeal mucosa	15
Orthopedics Morphix suture anchor	16
Orthopedic casting	17
Orthodontic	18
Ophthalmic applications	19
Novel McKibben artificial muscles	20
Micro-valves in micro gadgets	21
Medical micro-tweezers	22
Kidney dialysis needles	23
Treatment of hair	24
Manipulation and capturing of cells	25
Mending of cardiac valve	26
Fillers for bone defect	27
Aneurysm occlusion gadgets	28
Clot elimination gadgets	29
Controlled drug release	30
Endoscopic surgery suture	31
Bio-MEMs	32

Table 2. Textiles applications of shape memory polymeric textiles nanoarchitectures.[31–41]

Functional Textiles Applications	Ref
Wrinkle free finishing of cotton fabrics	33
Shape memory fibers	34
Shape changing nanofibers	35
Self-peeling dry adhesive	36
Pressure garments	37
Phase change fabrics	38
Memory foam mattress, pillow and insoles	39
Fashion design	40
Deodorant fabrics	41
Damping materials	42
Crease and pattern retention finishing	43
Damping fabrics	44



Scheme 1. General elucidation of the molecular architecture of SMPs composed of switching phases along with net-points[42] .

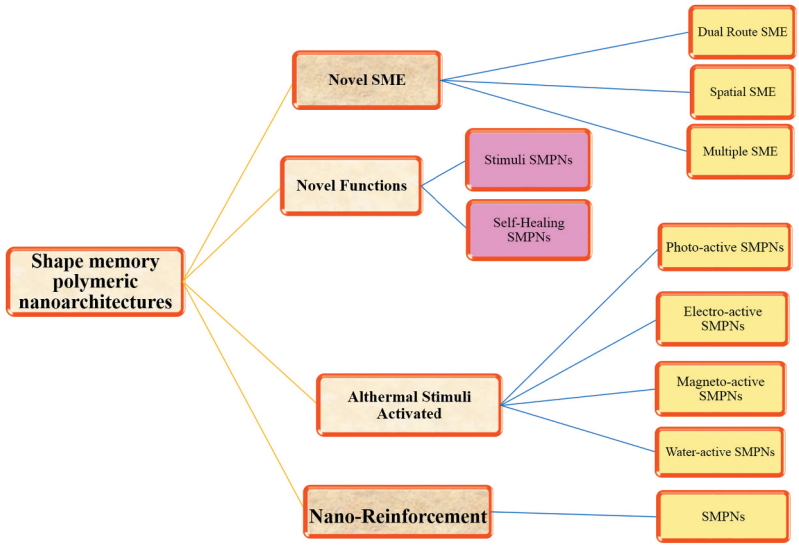


Figure 2. Achievements of SMPNs.

effects thereby inculcating novel as well as emerging functions as depicted in Figure 2.

From Figure 2, instances of athermal stimuli responsivity include electroactivity, magneto-activity, water-activity, and photo-activity. Furthermore, novel SMEs include multiple-SME, spatially controlled SME, and dual route SME. New functionalities include stimuli SMEs like magnetic field-ME, and self-repairing effect of SMPNs such as thermoplastic nanoparticulates embedded SMPs.[42]

Therefore, this paper elucidates recently emerging trends in construction, characterization, properties and multifunctional applications of SMPNs.

2. SMPNs nanoreinforcements

SMPs exhibit inferior intrinsically poor mechanical strength as well as shape recovery stress. These challenges have expansively limited the SMPs applications. Reinforcement nanofillers are capable of enhancing the mechanical behavior and shape recovery stress of SMPs via physically affiliated blending, chemical cross-linking, as well as in-situ polymerization.[43–45]

2.1. Carbon nanotubes (CNT) reinforced SMPNs and carbon nanofibers (CNFs) SMPNs

A major aim of CNT incorporation within SMP entails electroactive SME attainment via Joule heating in intrinsically affiliated thermally-responsive SMPs.[46] Notable challenges envisaged in CNT/SMP nanoarchitectures involves uniform dispersion of CNTs within the SMP matrix. Numerous strategies have been utilized including melt mixing,[21] in-situ polymerization, chaotic mixing subsequented by in-situ polymerization, solution mixing subsequented by casting, solution mixing facilitated by ultrasonic dispersion, as well as CNT chemical functionalization subsequented by cross-linking reaction. Chemically functionalizing CNT, is an effectual strategy, as chemically functionalization has the propensity of increasing the compatibility between CNTs and SMP matrices.[47] The fabrication of CNT@polymeric nanoarchitectures is generally conducted using varying strategies (direct, solution and melt-mixing as well as in-situ polymerization. Using conventional steps (extruding or injection molding, available polymeric matrix is prepared into its initial, permanent configuration B. This is subsequently followed by a procedure referred as programming, where the polymeric material undergoes deformation and fixation into the temporary configuration.[48]

Electrically conducting polymeric nanoarchitectures are amongst most investigated SMPNs, and can be fabricated through the inclusion of graphene, carbon nanofibers (CNFs), carbon black (CB), carbon nano-paper, single-walled carbon nanotubes (CNT), multi-wall carbon nanotubes (MWCNTs), and aligned conducting carbon-oriented nanoreinforcements.[49] The major function of these nanoreinforcements involves the conversion of electrical current into heat via Joule effect, SMPs so-actuating. CNT are amongst the most potential reinforcing materials for the fabrication of elevated performance multifunctional nanoarchitectures.[50] CNTs additionally display elevated flexibility in comparison to traditional fiber reinforcements.[51] The idea of electrically conducting SMPNs composed of MWCNT offer several challenges concerning good dispersion of the nanoreinforcement within the polymeric matrix, interactivity with the polymeric chains, establishment of electrically conductive

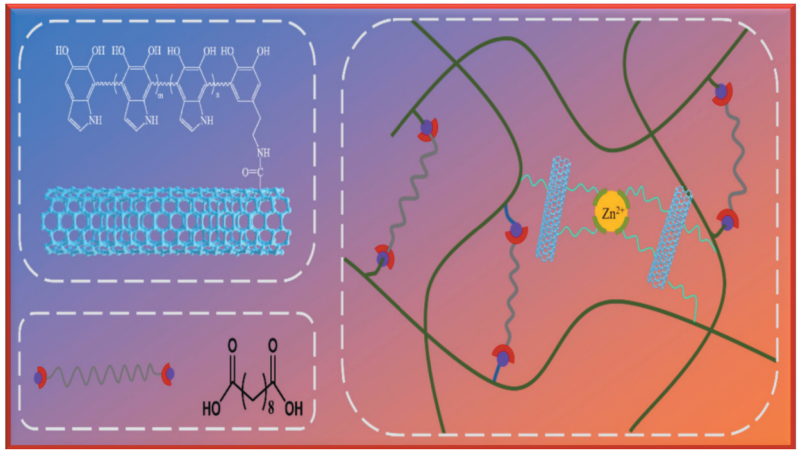


Figure 3. Chemical architectures of MWCNTs@PDA@epoxy nanoarchitectures in cross-linking by dynamical trans-esterification as well as metallic–catechol co-ordination.[59]

network within nanoarchitectures, and so on. These challenges impact directly on the end mechanical and thermal attributes of the materials, thereby inhibiting their applications. In a bid to mitigate these challenges and find panacea, numerous strategies have evolved including direct combination of MWCNT with the polymeric matrices (polyurethanes, polystyrene, and acrylates, epoxy and so on), surface functionalization of MWCNT and mixing with the polymeric matrix relative to enhancement of the interfacial bonding with the polymeric macromolecules, cross-linking (ionizing radiation vis-a-viz: electron beam or gamma radiation), the MWCNT with the polymeric matrices, along with MWCNT alignment with the polymeric matrices on applied electric/magnetic field or MWCNT conversion into nano-paper or film and its subsequent inclusion within the polymeric matrix[52–54].

Conventionally, the direct combination strategy of MWCNTs with epoxy matrix result in agglomerates formation and portray a self-supportive architecture of entangled CNTs in an irregular manner which are knitted together by Van der Waals interactions at the tube-tube junctions. Similar designs are usually utilized as fire along with lightning strike protection as well as electromagnetic shielding interference (EMI).[55,56]

In a work, an effectual strategy was evolved to enhance the interfacial interactivity between MWCNT/epoxy matrix.[57] The behavior of heat conductivity as well as strength of the epoxy vitrimer were improved through inclusion of polydopamine (PDA) coating. PDA is a prevalently utilized photo-thermal agent, efficient in functionalizing MWCNTs utilized in photo-responsive epoxy resin.[57] Vitrimers are a novel set of cross-linked polymeric materials, combining the features of both thermosets and thermoplastics.[58] Vitrimers are polymeric materials exhibiting cross-linking with dynamically exchangeable covalent bonding capable of being thermally rearranged while sustaining the integrity of the cross-linked architecture.[59] Hence, they can undergo repeated processing as thermoplastic substrates at a temperature beyond the topological-freezing transitional temperature (Tv). Figure 3 presents the chemical architectures of MWCNTs@PDA@epoxy nanoarchitectures in cross-linkage via catechol – metallic co-ordination as well as dynamic transesterification.[59]

Furthermore, the influence of MWCNTs@PDA on thermal conductivity, morphology, strength and stress relaxation, of the epoxy architectures were evaluated.[59] Additionally, PDA was revealed an effective route of mitigating the challenge of interfacial interactions between the MWCNTs and epoxy matrix attributed to efficient promotion of epoxy crystallization. The level of MWCNTs and MWCNT@PDA@epoxy nanoarchitectural distribution are presented in Figure 4A. From the fractured surfaces SEM images in Figure 4B, pristine epoxy displayed a smooth, thin fractured surface, with a brittle fractured behavior.[59]

The self-repairing, shape memory and recyclability of epoxy vitrimers is caused by stress relaxation induced by the dynamic trans-esterification exchange reaction. Figure 5A depict the classical epoxy/carboxylic acid polymeric architecture, the prevalence of both the free hydroxyl functional entities as well as the carboxylic ester functional entities.[59]

Figure 5B present the shape memory behavior of EP-M@PDA-1.0% using strip, spiral, and u-type specimens. The original specimens were strip specimens, configured at 200 C for 2 h on exposure to external force. Here, the strip specimens underwent deformation to a permanent spiral configuration at 70 C, beyond the glass transition temperature, with cooling to room

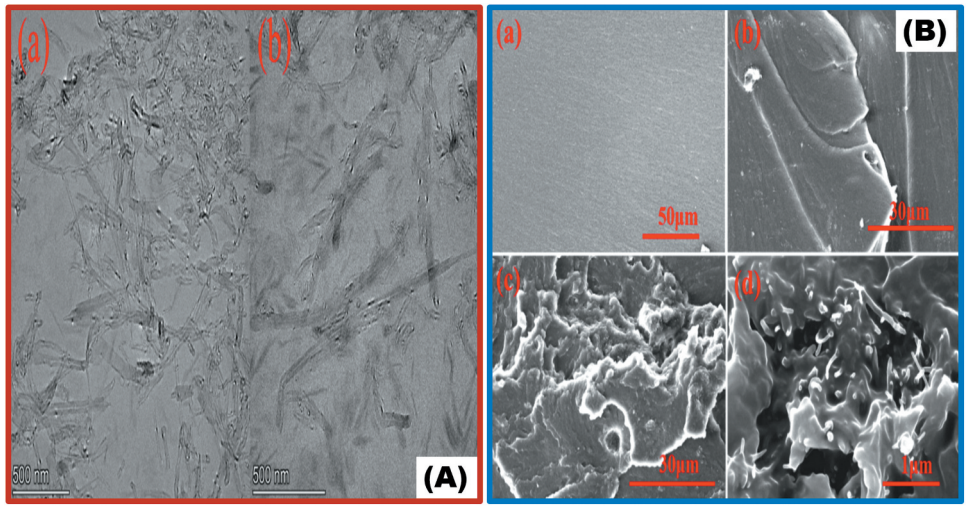


Figure 4. TEM images of (a) EP-M-1.0%. (b) EP-M@PDA-1.0%. B. SEM images of fractured surfaces for (a) EP. (b) EP-M-3.0%. (c) and (d) EP-M@PDA-3%.[59]

temperature. Post heating at 70 C, the configuration of the permanent spiral configuration recovered back to a strip configuration.[59]

Similar to CNT, CNFs exhibit outstanding electrical (106 S/m) and thermal conductivity (1000 W/mK).[60] Generally, to achieve improved interfacial bonding, they undergo oxidization and inclusion within the polymeric material through direct blending in form of a nanopaper or hybrid nanoreinforcement of CNF.[61] This facilitate the electrical actuation of SMPNs, presenting improved mechanical behavior.[62] Enhanced CNFs distribution can be attained via high-powered sonication or via in situ polymerization and subsequent lamination of the CNF papers upon styrene-oriented SMPs activated by an electric voltage.[63]

A work investigated feasibility of utilizing in situ generated hybrid polymer-polymer nanoarchitectures as polymeric materials exhibiting triple shape memory (TSM), which, unlike conventional polymeric blends with TSM, are characterized by completely separated segments transitional temperatures with strongest bonding between the polymeric blends segment interfaces which are imperative to shape fixation as well as recovery.[64] This was actualized utilizing three-component structured polylactide@polybutylene adipateterephthalate@cellulose nanofibers (PLA@PBAT@CNFs).[64] The function of in situ garnered PBAT nanofibers and CNFs during formation of effective physically affiliated cross-linkage at PLA@PBAT, PLA@CNF and PBAT@CNF interfaces and the influence of CNFs on PBAT fibrillation and crystallization procedures were examined.[64] Here, the in situ garnered SMPNs exhibited drastically elevated parameters of strain recovery ratios, strain fixity ratios, faster recovery rate and enhanced mechanical attributes in comparison with the pristine blend.[64]

The morphological behavior of PLA@PBAT blends as well as in situ achieved nanoarchitectures filled with CNFs was examined using SEM evaluation. The SEM images of the PLA/PBAT cryo-fractured surface of the blends and composites filled with 3 and 7 wt.% CNFs are presented in Figure 6. It is observed that the blends are characterized by a matrix-droplet architecture (Figure 6 a, b), while in situ achieved nanoarchitectures exhibit a fibril-matrix morphological architecture (Figure 6 c, d).[64]

Hence, in a work, CNTs@PDA nanoarchitectures were constructed via a nil-covalent bonding strategy in synergy with TPI through a melt-blending technique for fabricating novel SMP nanoarchitectures.[65] SEM results reveal that PDA nanoparticulates evenly decorated CNTs surfaces. From results garnered from DSC, TGA, and XRD, crystallization features, as well as the specimens' thermal stability composed of CNTs@PDA were enhanced, in comparison with those constituted of CNTs. Furthermore, the surface and fracture morphologies of the specimen were SEM examined, and results reveal that PDA remarkably enhanced the interfacial compatibility between CNTs@TPI, which further enhanced the mechanical features of the composites. Simultaneously, the CNTs@PDA (2.4 phr) nanocomposite displayed the best mechanical features as well as shape-memory behavior.[65] From this image, the surface of un-functionalized CNTs was somewhat smooth

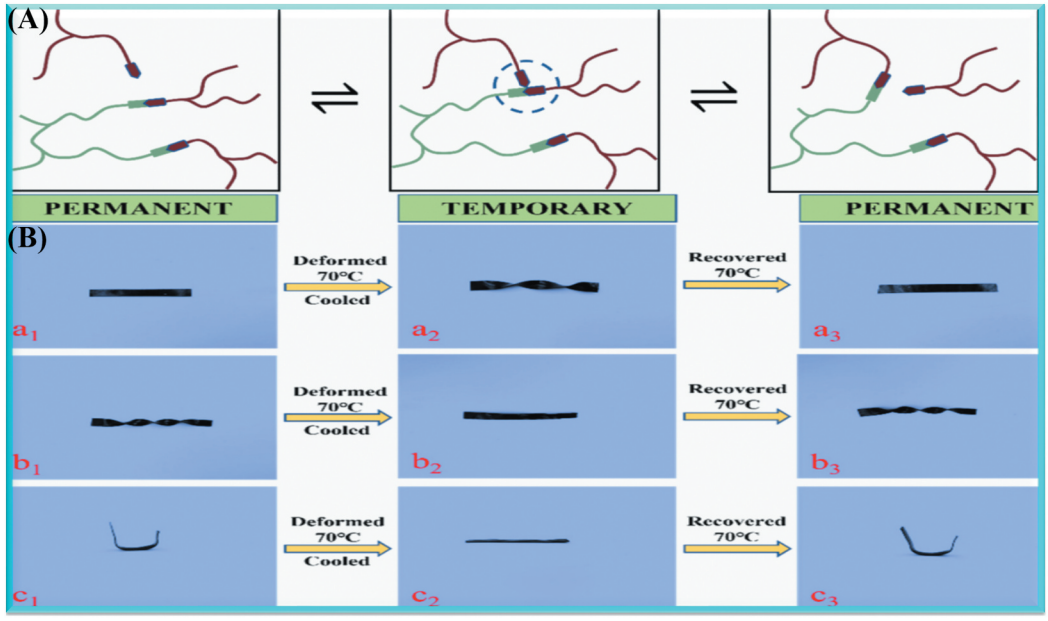


Figure 5. A. Epoxy vitrimers cross-linked by dynamic trans-esterification reaction. B. Shape memory disposition of (a1 – a3) strip specimen. (b1 – b3) spiral specimen (c1 –c3) u-form specimen.[59]

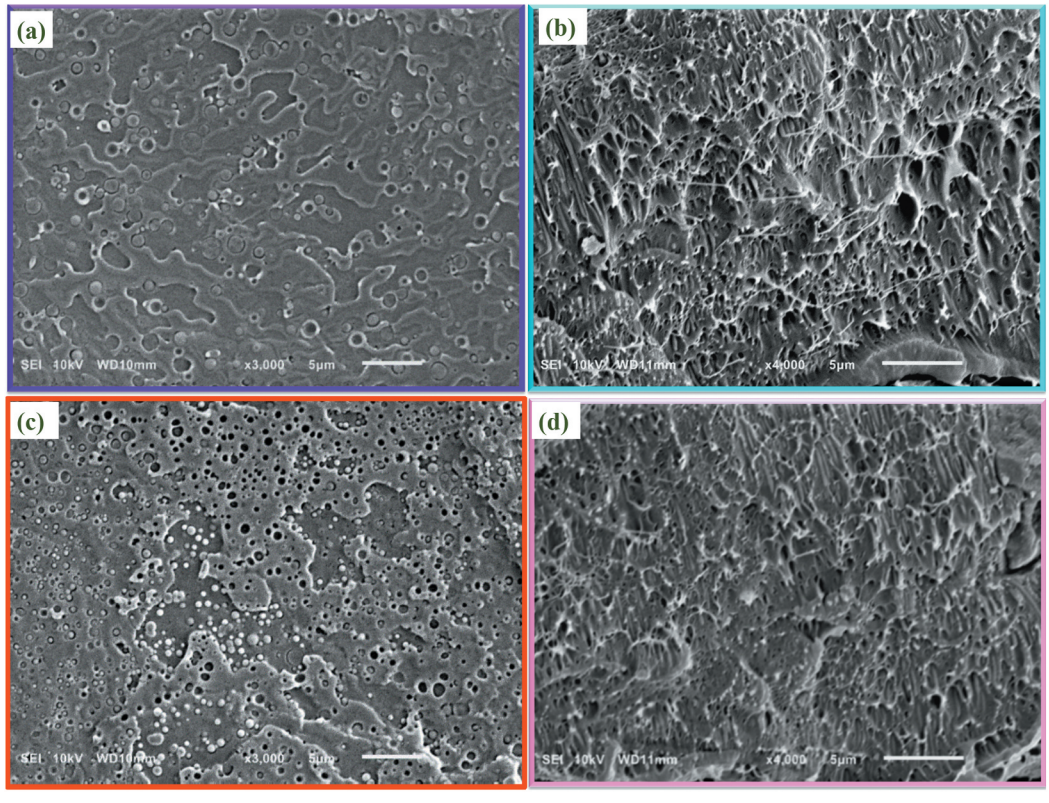


Figure 6. SEM images of cryo-fractured surfaces of PLA@PBAT@CNFs blends (a, b) and in situ generated composites (c, d). (a, c)—3wt. % CNFs, (b, d) —7wt. % CNFs.[64]

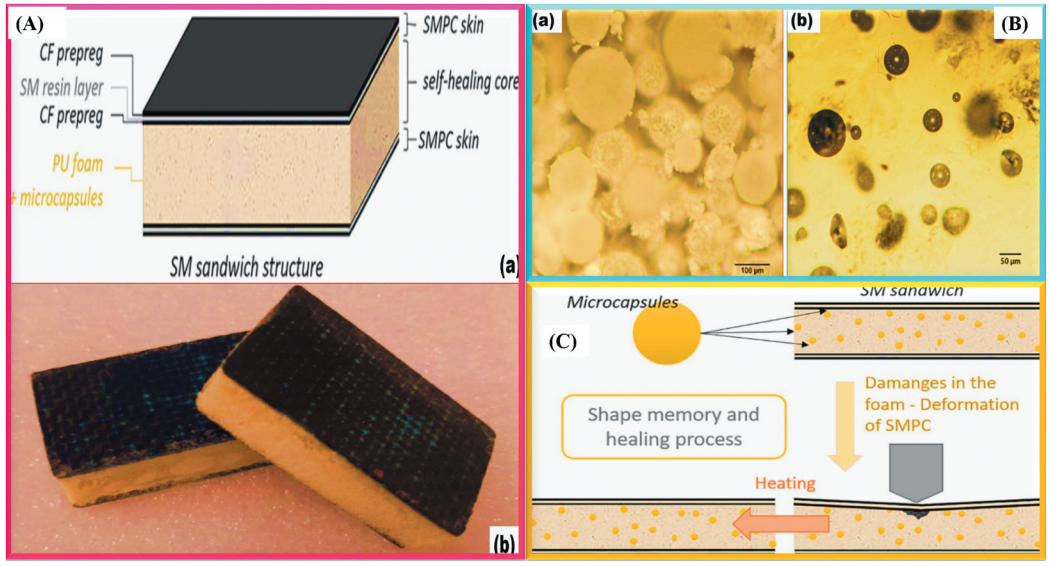


Figure 7. A (a) SM sandwich architecture; (b) image of the constructed sandwich specimen. B (a) Post-fabrication optical microscopic image of SH capsules; (b) image SH capsules incorporated within water-clear epoxy resin. C. Schematization of the shape memory and mending procedure.[65]

and clarified in the outlay, as the CNTs@PDA surface displayed an obvious coating sheet which was dense and comprehensive in architecture.[65]

The shape memory (SM) sandwich architectures were constructed utilizing two SMPC specimens of 100 30 mm^2 as outerlayers and the PU foam with and without microcapsules as the core of the architectures (Figure 7a,b).[65] This foaming volume facilitates attainment of a foaming ratio of about 4.5. The rate of shape recovery of each specimen was examined 20 times, thereby proving the performance stability of the SMPC. This sandwich architecture is schematized as shown in Figure 7a. Two specimen for individual condition, with and without microcapsules within the foam's core, were constructed (Figure 7b). Optical microscopic examination of the constructed SH capsules set was conducted to ascertain effectiveness of the procedure and determine the average size of the capsules (Figure 7a,b). The self-mending capabilities of the constructed SM sandwich architectures were ascertained utilizing a three-point bending test via a universal testing machine (MTS Insight 5) as presented in the schematization in Figure 7C.

Micrographic images of the PU foams with and without microcapsules are presented in Figure 6. The PU foam displayed a closed cells architecture wherein microcapsules of a darker color with sizes of varying micrometers are dispersed, as depicted in Figure 8A (a-d). The mending effect was also seen through micrographic images of deteriorated regions both in pre-mending (compressed specimen) and post-mending (post heating), as presented in Figure 8B (a-d).[65]

Shape fixity measured values and shape recovery are presented in Figure 9a. Irrespective of microcapsules prevalence, garnered results demonstrate that the PU foam exhibit shape recovery capability of about 94% and up to 79% fixity. Examination of the SME of the sandwich architectures, as depicted in Figure 9b, demonstrate how shape fixity as well as shape recovery are similar for all specimens, irrespective of microcapsules prevalence.[65]

Finally, the capability of designing and optimizing self-mending shape memory architectures at a macroscopic instead of molecular or micrometric level presents these structures appropriate for large-scale fabrication in versatile application areas, including aeronautics and automotives.[65]

In a related work, a number of polylactic acid/thermoplastic polyurethane (PLA@TPU) blends in synergy with multi-walled carbon-nanotubes (MWCNTs) were fabricated via extrusion for 3D printing, whilst the printed specimens were deeply investigated for their thermally-configured SME and mechanical features.[66] Initially, the morphological examination revealed that the blends were immiscible, as MWCNTs mixed with TPU phase, thus enhancing their interfacial adherence while improving the shape recovery response. Later, the mechanical properties (tensile strength and Young's modulus) exhibited descending trends after embedment of MWCNTs. Contrastingly, in wave architectured (3D-

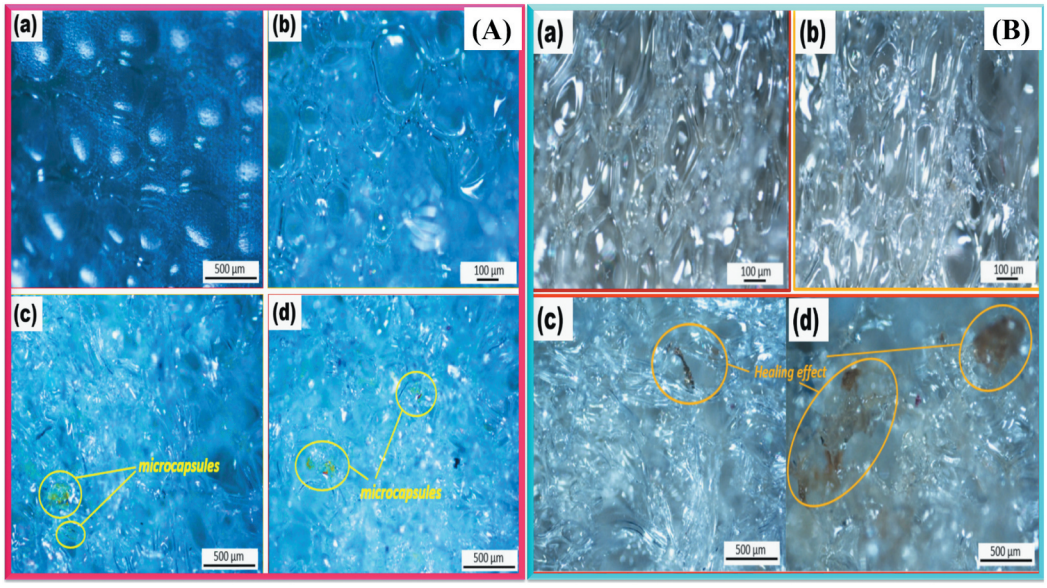


Figure 8. A. (a, b) optical microscopic images of PU foam devoid of microcapsules; (c, d) images of self-mending capsules within the PU foam. B. (a, b) optical microscopic images of PU foam post compression (shape fixity) prior recovery; (c) images of PU foam post recovery (mending effect).[65]

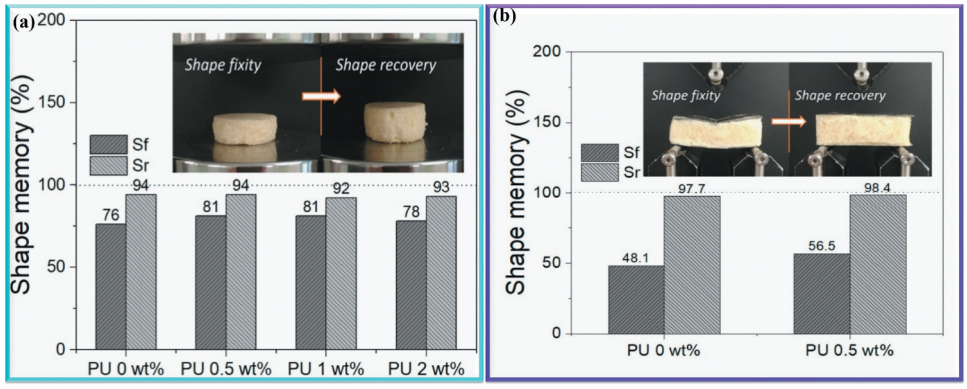


Figure 9. PU cylindrical foams shape memory behavior with and without microcapsules (a). (b) SM sandwiches load – displacement curves at initial state and post recovery.[65]

printed) specimens, a systematic increment in tensile strength along with Young's modulus was ascribed to the wave-length, in alliance with development of the shape-memory behavior (both recovery and fixity) as depicted in Figure 10.[66]

The wave architectured PLA@TPU@CNT nanoarchitectures show potential of alleviating shape-memory performance as well as dimensional stability for multifunctional applications.[66]

In similar investigation, PCL-TPU/MWCNT nanofibrous-architecture has been fabricated to function as outstanding sensing entity for wearable gadgets,[67] whereas in another work, polyolefin elastomer (POE)/lauric acid (LA)/carbon black (CB) referred qs POE/LA/CB nanoarchitectures exhibiting triple-stimuli responsive shape memory effect (SME) was demonstrated to be safe for a vast range of applications.[68]

2.2. SMPNs composed of cellulose nanocrystals

Ascribed to their renewability, stiffness, and biocompatibility, cellulose nanocrystals CNCs, previously termed cellulose nanowhiskers, have demonstrated suitability as nanofiller for varieties of biopolymeric materials.[69] Furthermore, CNCs are garnered from very abundant biopolymeric materials which offer numerous benefits, including availability, inexpensiveness, low density, renewability along with chemical and physical features.[70] Their chemical architecture in the presence of hydroxyl entities offers feasibility of forming

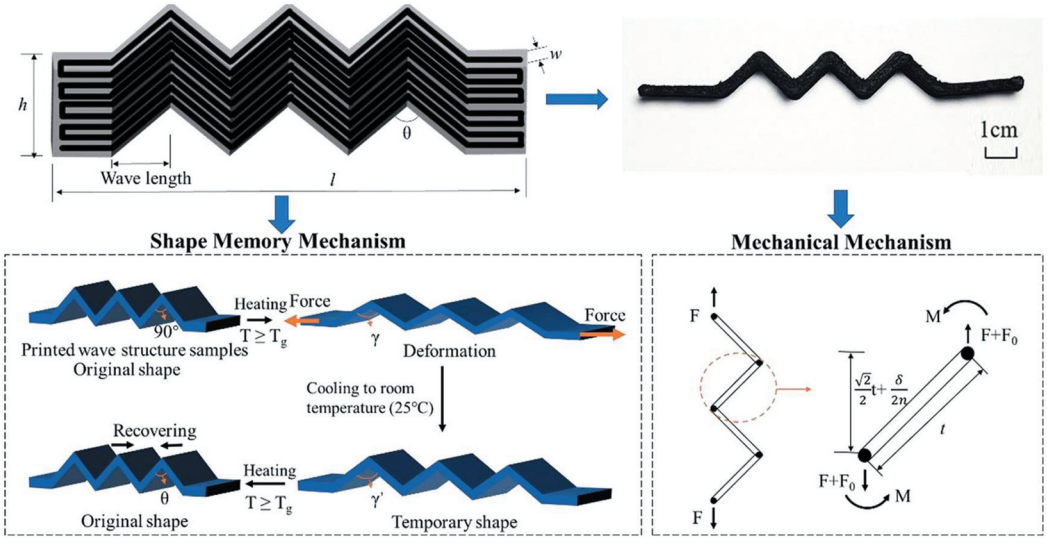


Figure 10. Shape memory effect of PLA@TPU@CNT nanoarchitectures.[66]

elevated-modulus and interconnectivity of CNCs architecture within the polymeric matrix via hydrogen bonding. An advantage of this nanoreinforcement is that display of elevated stiffness and high aspect ratio.[70]

Generally, CNCs are possesses low toxicity, presenting them attractive for fabrication of water-responsive SMCs with prospects for biomedical applications (self-tightening sutures, self-retractable as well as removable vascular stents).[71] Nevertheless, the fabrication of nanoarchitectures through direct physical encapsulation of CNCs within polymeric matrices is challenging because of the inferior affinity between the nanoreinforcement and the polymeric chains resulting in insignificant increment in or even nil improvement of the mechanical features of the polymeric materials. On incorporation of cyclodextrin (CD) within SMPs, enchanting and outstanding functionalities are inculcated thereby broadening their prospective applications.[72]

Shape memory polyurethane (SMPU) has been versatily perceived as prospective smart materials, whereas, porous SMPU exhibited inferior shape memory along with mechanical features. Hence, in a work, nanocellulose was embedded within SMPU to enhance all the properties of the porous SMPU via 3-D printing prototyping as elucidated in Figure 11. Results affirmed the potential applications of SMPU/CNC nanoarchitectures for smart biomaterials.[73]

In another work, CNC@SMP bionanoarchitectures were constructed via in situ single-route procedure as depicted in Figure 12.[74]

The inclusion of about 10 wt. % CNCs induced a notable improvement in the tensile strength at yield and modulus of elasticity while maintaining the elongation at break, ascribed to the combined activities of CNCs as a nucleating entity for crystallization and elevated compatibilization of the reinforcement agent of the architecture. Furthermore, the in situ embedment of CNCs improved the shape memory ability of polyurethanes, thereby enabling its usage in functional material applications, including the biomedical field.[74]

Another work demonstrated outstanding thermal, mechanical, shape memory, and self-mending properties of 3-D printable, thermos-reversible, cross-linked network architecture.[46] Here, the thermos-reversible cross-linking route is constituted of Diels-Alder (DA) reversible covalent and transient cross-linkage, fabricated utilizing thermoplastic polyurethane (TPU) and poly(ε-caprolactone) (PCL) with cellulose nanocrystalline (CNC) as the cross-linking entity. The inclusion of CNC, functionalized using furan (CNC-FA) and maleic anhydride groups (CNC-MAH), exhibited outstanding compatibility with TPU@PCL matrix, resulting in formation of physically inclined and chemically affiliated cross-linking architectures in the nanoarchitectures, as depicted in Figure 13.[46]

Additionally, the mechanical, thermal, and self-mending features of the material were significantly enhanced after the inclusion of functionalized CNC.[46] The mechanically stabilized 3-D printable self-mending nanoarchitectures facilitate the construction of lasting conductive gadgets and biomimetically affiliated skin gadgets with elevated mechanical features, outstanding electrical mending and superior strain-propagated fluorescence features, exhibiting prospects in next-generation flexy electronics, encryption gadgets as well as electronic skins, thereby expanding applications of 3D-printable materials.[46]

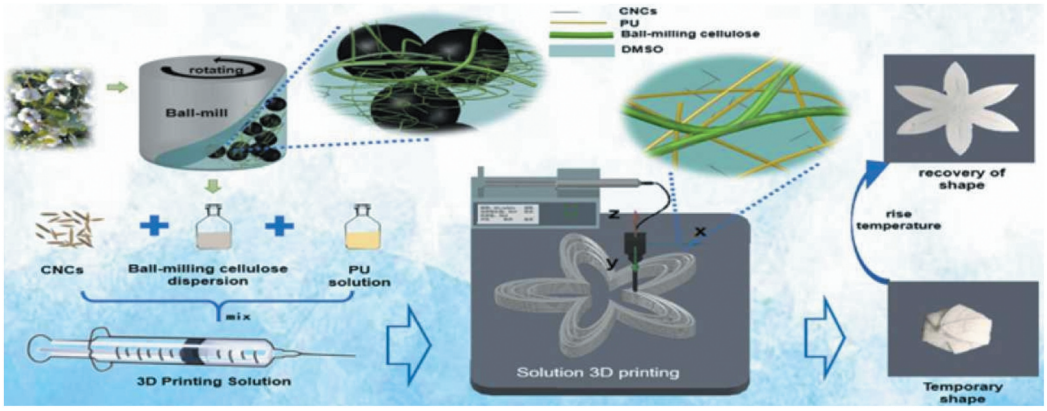


Figure 11. Construction, 3-D prototyping, and shape memory behavior of SMPU/CNC nanoarchitectures.[73]

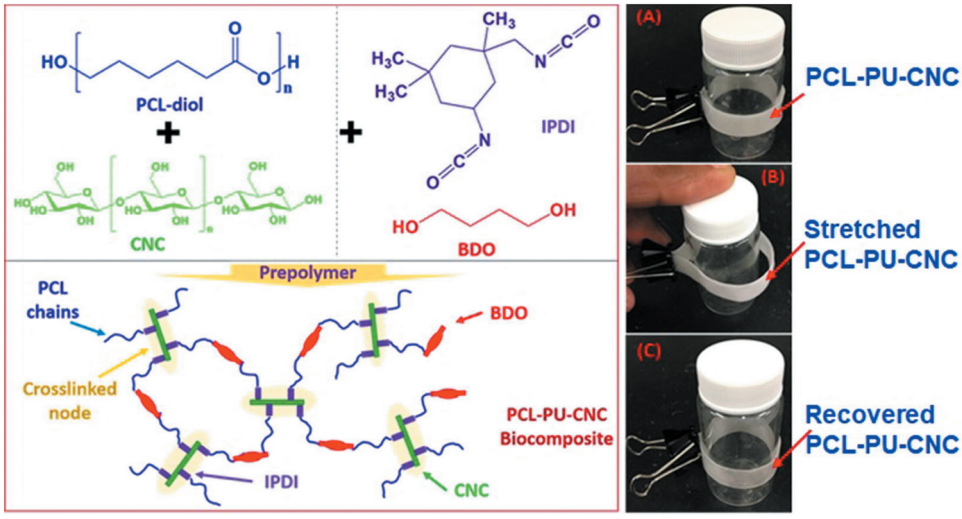


Figure 12. Construction route, characterization and shape memory behavior of PU@PCL@CNC bionanoarchitectures.[74]

In another investigation, thermoresponsive and water-responsive shape-memory polymeric nanoarchitectures were constructed via chemical cross-linkage of cellulose nanocrystals (CNCs) with polycaprolactone (PCL) and polyethylene glycol (PEG) referred as PCL@PEG@CNC bionanoarchitecture as depicted in Figure 14.[75]

Hence, this thermoresponsive and water-responsive shape-memory nanoarchitectures could undergo potential developed into a novel smart biomaterial.[75] Hence, PCL@PEG@CNC bionanoarchitecture depicts a variety of applications as medical gadgets, drug delivery systems, anti-cancerous therapies, and biologically for hydrogels, bio-artificial organs, as well as tissue engineering. PCL@PEG@CNC bionanoarchitecture has potential use in the construction of 3-D scaffolds for bone tissue engineering attributable to advantages (biocompatibility, gradual rate of degradation, and load-carriability.[75]

In another investigation, a pH-responsive shape-memory polymeric nanoarchitecture was constructed through the combination of poly (ethylene glycol) @poly(ε-caprolactone)@polyurethane (PECU) with modified cellulose nanocrystals (CNCs). CNCs were functionalized with pyridine moieties (CNC – $C_6H_4NO_2$) via hydroxyl replacement of CNCs with pyridine-4-carbonyl chloride and with carboxyl groups (CNC – CO_2H) via 2,2,6,6-tetramethyl-1-piperidinyloxy (TEMPO) mediated surface oxidation, respectively.[6]

The shape memory behavior of this material only depended on environmental pH variation. Hence, this pH-responsive shape-memory nanoarchitecture potentially be constructed into a novel smart polymeric nanoarchitecture.[6]

In another investigation, a number of thermoplastic polyurethane (TPU)@carbomer (CB)@nano-celluloses (CNCs) multi-responsive shape-memory nanoarchitectures were

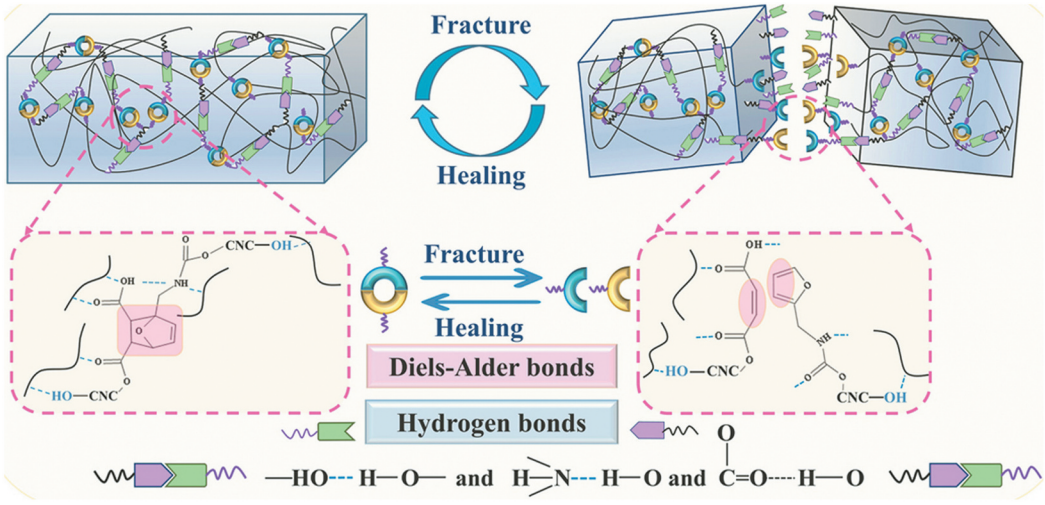


Figure 13. Fracture and healing mechanism of TPU@PCL@CNC-FA hybrid nanoarchitecture.[46]

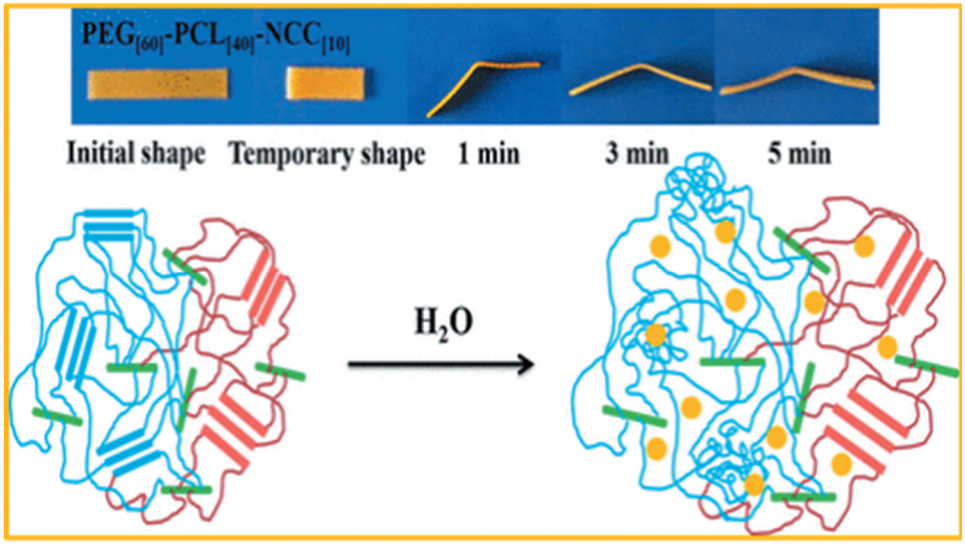


Figure 14. Construction of PCL@PEG@CNC bionanoarchitecture.[75]

customized utilizing CNCs as a cross-linking agent.[76] The effect of CNCs composition on the mechanical behavior of TPU@CB@CNC nanoarchitecture has been elaborately investigated with the best CNCs inclusion pegged at 5 wt. %. Results revealed, that the nanoarchitecture exhibited multi-responsivity for heat, water, pH, and ethanol as illustrated in Figure 15, demonstrating its prospect in slow-drug releasing, flexible robotic and electronic applications.[76]

Cellulose-oriented water-induced shape memory materials have garnered great attention because of their versatile range of sources, elevated rate of responding, and ecobenign disposition. Nevertheless, shape memory polymers (SMPs) with rapid responsivity tend to lose wet strength as a result of too much hydration tendencies, which highly restrains the application versatility of cellulose-oriented shape memory water-responsive materials. In order to mitigate this challenge, a work constructed, cellulose nanofiber (CNF)@polyvinyl alcohol (PVA)@lignin (LIG)@Citric acid (CA) referred as CA@LIG@PVA@CNF hybrid membranous nanoarchitecture with elevated water-responsive shape memory features as depicted in Figure 16.[77]

The CNF nanoarchitectural films exhibited outstanding performance at inhibiting ultraviolet as well as transmittance value of the membrane less than 8% in the ultraviolet region of 200–400 nm. Hence, the resultant CA@LIG@PVA@CNF membrane displayed potential usage in smart technology while offering a framework for constructing a cellulose-oriented shape memory polymeric nanoarchitecture.[77]

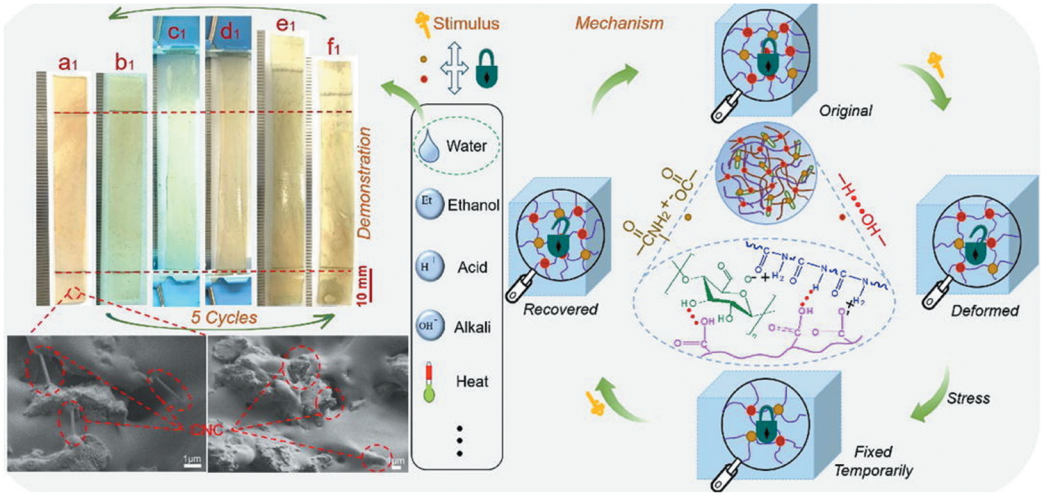


Figure 15. Shape memory responsivity of TPU@CB@CNC nanoarchitectures.[76]

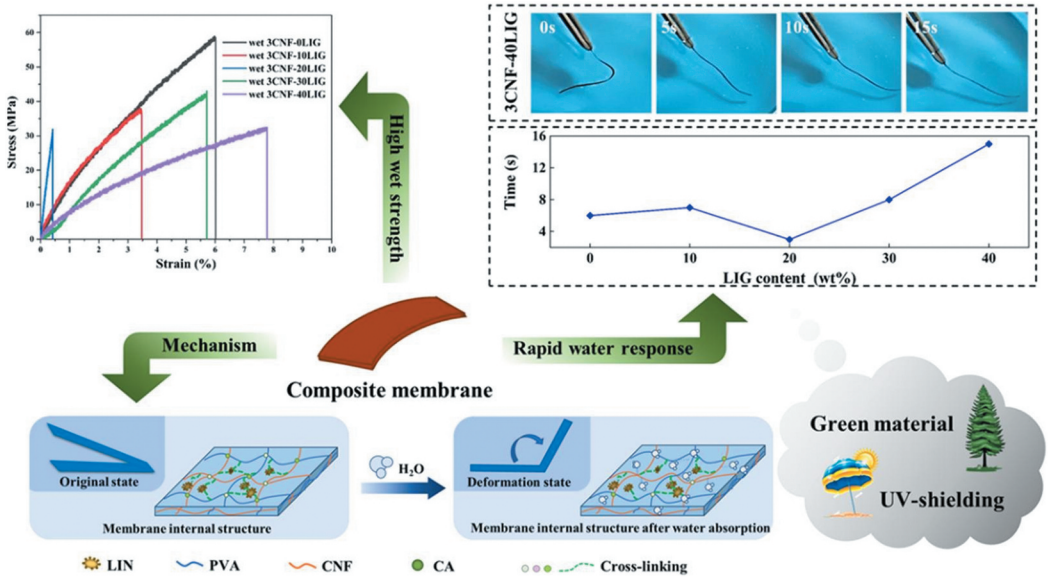


Figure 16. Mechanical features and water responsive shape memory recovery mechanism of CA@LIG@PVA@CNF hybrid membranous nanoarchitectures.[77]

2.3. Nanoclay shape memory polymeric nanoarchitectures

SMPNs are polymeric nanoarchitectures with the ability of returning to their earlier programmed configuration post exposure to external stimulus. Material properties enhancement with nanoclay reinforcement has improved its thermomechanical attributes while escalating its scope of industrial applications.[78]

The quest for novel types of actuating equipments have continually grown, and novel strategies have been made feasible by the emergence of novel materials and construction approaches. Self-propelled actuating equipments have garnered significant interests because of inherent susceptibility to be propelled by entities in ambient clime.[79] This form of actuating equipments can undergo utilization in flexible strain sensors, soft robotics, smart breathing textiles and artificial muscles.[80] Nevertheless, synthetic polymeric matrices are not ecobenign and induce ecological challenges. Biodegradable biopolymeric materials usage has become panacea to ecological challenges.

Polylactic acid (PLA) exhibit biodegradability and biocompatibility with elevated prospects. In a work, nanoclay filled PLA/PU nanoarchitectures yarn was constructed with high twist.[81] The twisted yarn then underwent shaping to a coiled architecture via mandrel

annealing. Results revealed that the nanoarchitecture yarn exhibited a dual route shape-memory effect in a twisted-coiled architecture. It additionally revealed a remarkable reversible contraction stroke within low temperature range.[22]

Shape memory nanoarchitectures of PU@nanoclay were constructed via melt blending of PU@nanoclay. Relying on nano-indentation and micro-hardness examinations, the nanoarchitectures strength sharply escalated based on nanoclay composition, ascribed to the enhanced nanoclay – polymer interactions. Thermal mechanical examinations revealed good mechanical and shape memory impact of the nanoarchitectures. Complete shape memory recovery was exhibited by both pristine PU as well as PU-clay nanoarchitectures.[82] High resolution transmission electron microscopy (HR-TEM, JEOL 2010F) revealed that modified nanoclay formed a 3-D bundled network architecture wherein the length of single clay fiber changed from sub-micrometric level to a few micrometers whereas the diameter was in the order tens of nanometer (see Figure 17(a)). The ring-like scattered diffraction region expose the nanocrystalline attribute of the modified fibers as shown by the inset electron diffraction pattern in Figure 17(a). The corresponding HR-TEM images in Figure 17 (b, c) affirm that single crystallites are inculcated within amorphous matrix with isolation of a few nanometers.[82]

Figure 18a presents the heat-mechanical cyclic examination of the 30 wt.% nanoclay nanoarchitecture. A remarkable shape recovery was seen as the shape recovery rate was 99.2% in initial tensile cycle and 97% within the second round. The maximum stress reduced by 8% ascribable to some flaws garnered whereas creep took place during continual loading at an air temperature of 60°C.[82]

In a bid to expose the shape memory effect, thin beams of pristine PU and 30 wt.% nanoclay/PS nanoarchitecture, possessing cross sectional area (c.s.a) of 2×2 mm^2, were bent post-heating to 80°C, while the shape were fixed during cooling at room temperature (20°C). The shape recovery was demonstrated on a hotplate with a surface temperature of 80°C. The pristine PU specimen displayed a sharp response, recovering to its initial configuration within 30 s (see Figure 19 (c)). The nanoarchitecture beam with 30 wt.% nanoclay also exhibited a complete shape memory recovery with 60 s (see Figure 19 (d)). The slow recovery of the nanoarchitecture specimen is ascribed to the inclusion of nanoreinforcement inhibiting the molecular chains movement, such that the shape memory effect was mildly delayed. TEM image of nanoclay distribution within PU is presented in Figure 20.[82]

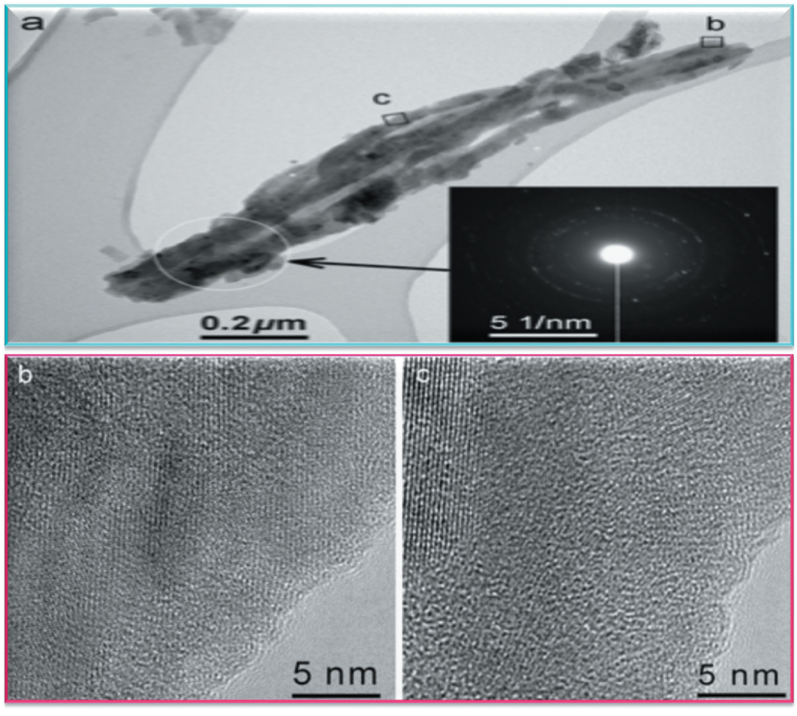


Figure 17. TEM images of thermally-modified nanoclay: (a) General and electron diffraction pattern, (b) and (c) HR-TEM micrographic images taken at specific positioning of nanoclay cluster.[82]

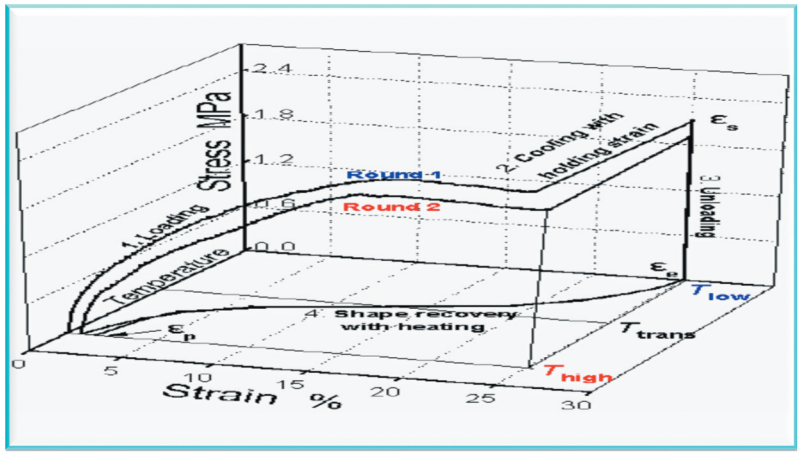


Figure 18. (A) Thermal cyclic tensile results of 30 wt.% nanoclay SMP nanoarchitecture.[82]

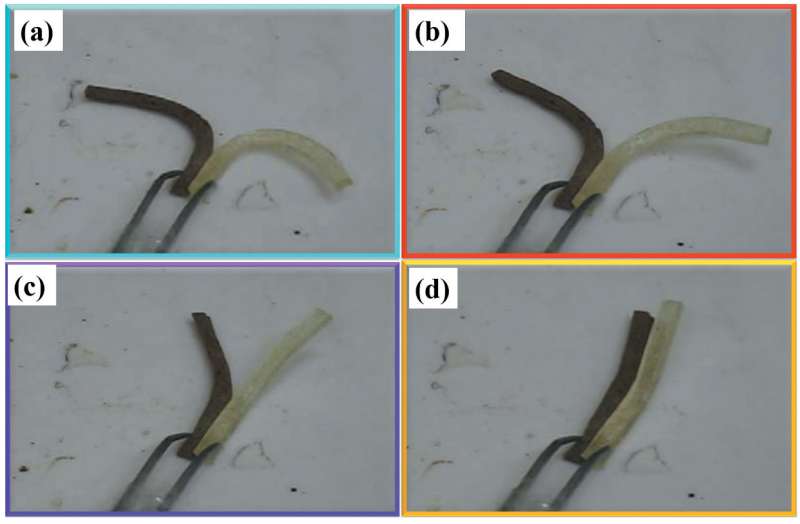


Figure 19. Recovery of shape memory specimens heated on a hotplate with a surface temperature of 80°C.[82]

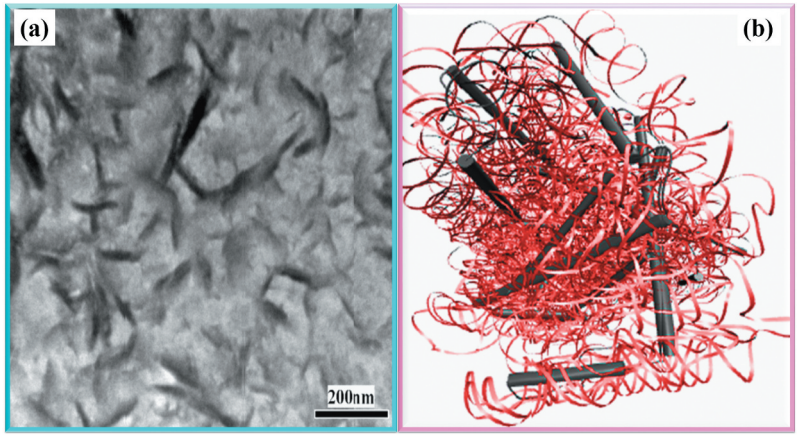


Figure 20. (A) TEM image of nanoclay. (b) Schematic elucidation of nanoclay distribution within the polymeric matrix.[82]

3. Challenges and future prospects

Despite the vast range of feasible geometry to adopt, key flaws of SMP includes poor stiffness and tensile strength, and general lack luster performances. Other notable limitations entail inferior heat dissipation, inertness to electromagnetic, light and electrical stimuli in along with poor sensitivity and inferior time of actuation recovery. Hence their potential uses are limited especially in regions where high performance is critical. Therefore, to find panacea to these challenges, a new class of shape-memory nanoarchitectures has sufficed. In the interim, numerous works have been conducted on shape memory polymer and nanoarchitectures recently with versatile enhanced properties for multifunctional applications[83–164]. On the other hand, polymeric nanoarchitectures have been constructed with superior features for multifunctional applications.[165–228] It is anticipated that the future of SMPNs is bright at all spheres of human endeavor.

4. Conclusion

In present review, the fundamental design and mechanisms of SMPNC have been presented elucidating the nanometric designing of nano-sized particulate-embedded SMP nanocomposites and applications. Specific examples were discussed to expose the underlying principles and emerging applications. In previous decades, a broad range of research activities have been conducted to garner novel SMPN while enhancing the available ones for elevated performance. Nevertheless, some emerging SMP phenomena have evolved which improve the flexibility of existing shape memory technology, while additionally exploring new horizons for enhanced versatility as well as adoptability. Thus, a range of emerging concepts have displayed potential for a versatile range of engineering applications. Therefore, the future of stimuli-responsive shape memory polymeric nanoarchitectures is anticipatedly blue ship.

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