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# Applications and Mechanistic Insights Into Intrinsically Self-Healing Polymers With Multifunctional 2D Materials

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## ABSTRACT

Self-healing (SH) polymeric composites hold the promise of revolutionizing material performance and durability, but the challenge lies in achieving a delicate balance between healing efficiency and mechanical strength. Healing processes typically require dynamic, reversible bonds, which can weaken overall material strength, whereas robust materials rely on strong covalent bonds that resist healing. 2D materials offer a solution by acting as nanofillers that not only improve mechanical properties but also introduce multifunctional benefits like electrical and thermal conductivity, responsiveness to stimuli, and enhanced barrier properties. Depending on their surface chemistry, these materials can either actively participate in the healing process or passively reinforce the polymer matrix. This review examines recent advancements in SH polymer composites enhanced with 2D fillers, exploring how factors like filler type, surface interactions, and loading levels impact both healing efficiency and mechanical strength. It compares the contributions of various 2D materials, identifying similarities and critical differences in their roles within polymer matrices. The article also highlights the need for standardized testing and advanced characterization techniques to better understand interfacial properties and healing mechanisms. By addressing current knowledge gaps and proposing future research directions, this review provides a comprehensive resource for advancing SH polymer systems, particularly in the integration of 2D materials for applications ranging from aerospace to electronics.

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**Abbreviations:** AA, acrylic acid; AFM, atomic force microscopy; AM, acrylamide; ANF, aramid nanofibers; APTES, (3-aminopropyl) triethoxysilane; BC, bacterial cellulose; BDS, broadband dielectric spectroscopy; BP, black phosphorus; CFRP, carbon fiber reinforced polymer; CMC, carboxymethyl cellulose; CNF, cellulose nanofibers; CNT, carbon nanotubes; CTH, close-then-heal; CVD, chemical vapor deposition; DADiels, Alder; DGEBA, bisphenol A diglycidyl ether; DMA, *N*,*N*-dimethylacrylamide; DMAEMA2, (dimethylamino)ethyl methacrylate; DTT, dithiothreitol; EMI, electromagnetic interference; EPR, electron paramagnetic resonance; FT-IR, Fourier transform infrared spectroscopy; GFRP, glass fiber reinforced plastic; GNP, graphene nanoplatelets; GO, graphene oxide; HEMA2, hydroxyethyl methacrylate; IR, infrared; LDH, layered double hydroxide; MAS, magic angle spinning; MAUPy2, (3-(6-methyl-4-oxo-1,2,3,4-tetrahydropyrimidin-2-yl)ureido)ethyl methacrylate; MEA2, methoxyethyl acrylate; MMP, *N*-methoxymethyl acrylamide; MMT, montmorillonite; MPTC, [3-(methacryloylamino)propyl]trimethylammonium; MSC, microsupercapacitor; NaSS, sodium *p*-styrenesulfonate; NDT, non-destructive testing; NOESY, nuclear Overhauser effect spectroscopy; NR, natural rubber; ODA, *n*-octadecyl acrylate; P4VP, poly(4-vinylpyridine); PAA, polyacrylic acid; PANI, polyaniline; PAS, positron annihilation spectroscopy; PBA2, [[(butylamino)carbonyl]oxy]ethyl ester; PBS, polyborosiloxane; PCL, polycaprolactone; PDMA, poly(*N*,*N*-dimethylacrylamide); PDMS, polydimethylsiloxane; PEA, poly(propylene glycol) bis(2-aminopropyl ether); PEG, polyethylene glycol; PEI, polyethylenimine; PEVA, poly(ethylene-vinyl acetate); PMAEFc, poly(2-aryloyloxy)ether ferrocenecarboxylate; PMMA, poly(methyl methacrylate); PNC, polymer nanocomposites; PNIPAM, poly(*N*-isopropylacrylamide); POSS, polyhedral oligomeric silsesquioxane; PU, polyurethane; PUU, poly(urea–urethane); PVA, polyvinyl alcohol; rGO, reduced graphene oxide; SDS, sodium dodecyl sulfate; SEM, scanning electron microscopy; SH, self-healing; SMA, stearyl methacrylate; SMASH, shape-memory assisted self-healing; SPR, surface plasmon resonance; SSNMR, solid-state nuclear magnetic resonance; SVET, scanning vibrating electrode technique; TEM, transmission electron microscopy; TENG, triboelectric nanogenerator; TMD, transition metal dichalcogenide; UV–Vis–NIR, ultraviolet–visible–near infrared spectroscopy; $\beta$-CD$\beta$, cyclodextrin.

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## 1 | Introduction

The remarkable ability of living organisms of self-healing (SH) after physical damage has inspired extensive research in engineering materials with similar regenerative capabilities. Replicating nature's self-repair mechanisms in artificial materials holds tremendous promise, potentially revolutionizing their life cycle. This is highly driven by the need for multifunctional polymers with superior performance, more reliability, and lower environmental impact. Thermoplastics inherently are better for the environment, and they also possess other interesting properties like tactility, easy reprocessability, and lighter weight compared to alternative materials like metals, wood, or ceramics. However, common thermoplastics also have lower mechanical properties, lower creep resistance, and lower thermal stability. On the other hand, thermosets possess favorable mechanical properties, but they are difficult to recycle. This highlights the need for stronger counterparts, either thermoplastics or thermosets, that have enhanced durability and reconnected bonding mechanisms for better recycling, which is what is offered by SH polymers.

SH polymers offer the potential to restore the performance attributes of the material either partially or fully after sustaining damage. Damage, in this context, refers to a localized reduction in the material's performance compared to its overall integrity [1]. This could simply mean that the material has lost its initial mechanical strength. In many cases, these materials possess the capability to detect damage and initiate a localized repair process autonomously.

The concept of SH polymers was first mentioned in 1969 by Russian researchers, who highlighted the significance of temperature in SH processes and the influence of changes in the relaxation spectrum of poly(vinyl acetate) on crack healing [2]. In 1984, Ellul et al. demonstrated that good contact between the surfaces to be repaired was essential for effect healing and introduced the concept of self-adhesion, thereby introducing SH as we know today [3]. Subsequently, works were scarce on this topic until the early 1990s, when it picked up some momentum when the concept of autonomous SH was introduced by Dry et al. for cement and epoxy resins [4, 5]. However, it was not until the 2000s, with the introduction of the first SH polymers designed for engineering applications, that significant interest was sparked, leading to extensive research in this field [6–8]. Since then, research on SH polymers has surged, with the number of publications increasing each year, where over 1700 studies were published in 2024, according to Web of Science. This surge in research activity underscores the growing recognition of the potential applications where SH polymers can improve performance and drive sustainability. Chen et al. in 2002 developed an SH polymer utilizing multiple furan and maleimide monomers based on Diels–Alder (DA) chemistry, pioneering the work in DA chemistry for SH [9]. They conducted fracture toughness tests on the material after heating it to temperatures ranging from 120°C to 150°C, where it demonstrated a 57% efficiency in regaining its maximum load capacity. This study highlighted two important facts: (1) Pressure has negligible effect on the material's healing performance, and (2) multiple repairs are possible by demonstrating 80% recovery rate after three healing cycles. Later, Cordier et al. introduced the concept of intrinsic SH to an elastomer by creating molecules that could automatically rechain and crosslink via hydrogen bonds, forming a labile supramolecular network [10]. This network could self-repair at ambient temperatures. Their findings indicated a time-dependent healing process, with longer durations leading to higher efficiencies, and confirmed that SH was not immediate. Presently, seamless incorporation of healing agents or mechanisms into the polymer matrix without compromising the material's original mechanical properties or functionalities. Integration would ensure that the SH components are uniformly distributed and maintain compatibility with the host matrix, enabling consistent performance across the entire material.

Recent research has focused on incorporating nanofillers, particularly 2D materials, as a way to address integration challenges while improving the mechanical, thermal, and functional properties of SH polymers. Their large surface area, adaptable surface chemistry, and multifunctional characteristics make them especially effective in enhancing overall performance [11]. This comprehensive review focuses on intrinsically SH polymers augmented with diverse 2D materials as active fillers. Unlike previous studies centered on individual 2D materials, this review aims to provide a holistic understanding of their advantages and common challenges. Moreover, it delves into the nuanced effects of material type and surface chemistry concerning specific end-applications. Beyond summarizing recent advancements in SH polymer composites incorporating various 2D materials (as also shown in Figure 1), including graphene and those beyond, this review underscores the need for standardized testing, advanced material characterization, and exploration of interface properties using sophisticated monitoring techniques. By analyzing statistical data from the literature, this review also identifies emerging areas warranting attention from the scientific community and provides a deeper understanding of current challenges and future perspectives.

## 2 | Mechanistic Insights and Design Considerations

### 2.1 | Failure Trajectory for Engineering Polymers

SH is a critical concept in understanding the lifecycle of engineering polymer materials. When assessing a composite's lifecycle (as shown in Figure 2A), the key point of failure is fracture, as this marks the onset of performance degradation [12]. Beyond this stage, the material's properties progressively decline until they fall below the mechanical threshold required for reliability, necessitating its removal from service. Although mechanical reinforcement of the macromolecular structure can delay failure, it does not fundamentally alter the failure trajectory [13]. In contrast, SH materials can initiate a healing process immediately upon failure, aiming to restore the material to its original state. Ideally, an SH material would activate healing instantly, achieve full recovery within a short timeframe, and sustain multiple repair cycles [14]. However, in real-world polymers, the efficiency of healing depends on the nature of their chemical bonding. Polymers with strong covalent bonds may exhibit slower healing kinetics, yet they can achieve a more durable recovery, thereby extending the material's overall lifespan. Conversely, polymers

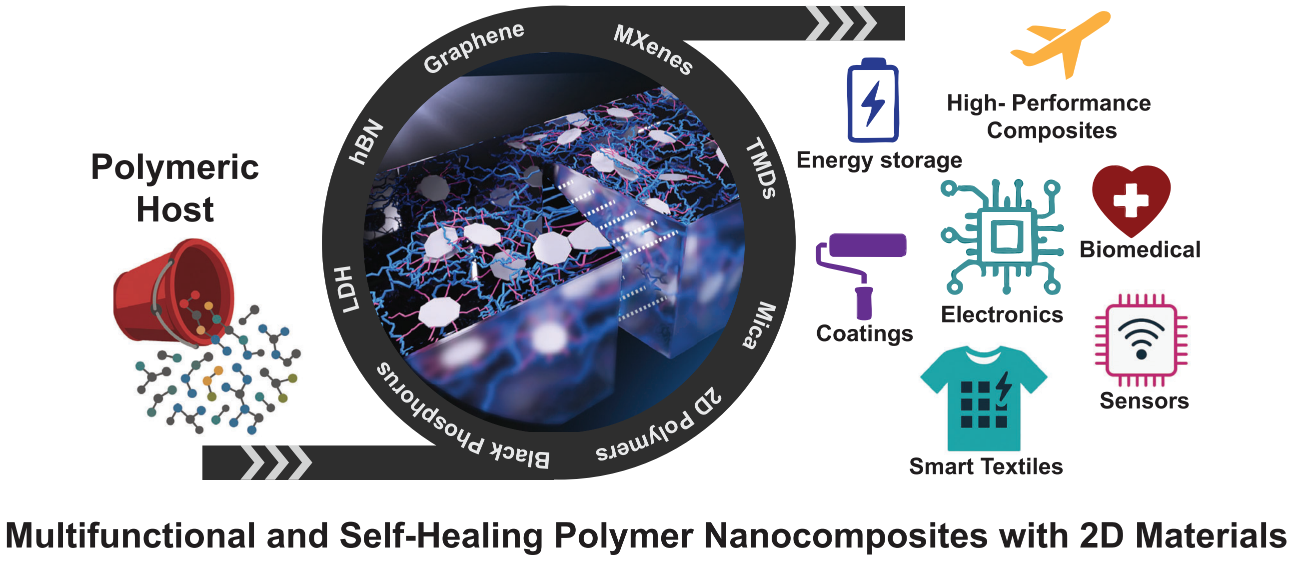

**FIGURE 1** | Overview of multifunctional self-healing polymer nanocomposites with 2D fillers and representative applications. LDH, layered double hydroxides; TMD, transition metal dichalcogenide.

with labile bonding mechanisms, such as hydrogen bonding, may heal rapidly but could also degrade more quickly under mechanical stress [15]. Regardless of these variations, incorporating SH functionality can effectively prolong the period during which the material remains above the minimum reliability threshold, thereby enhancing its service life.

## 2.2 | Underlying Mechanics

It is vital to understand the mechanics of the SH process in order to design efficient SH systems. Three key principles have been defined in the past to complete the SH process, namely, localization, temporality, and mobility, but now with modern multifunctional materials, it is essential to identify the fourth characteristic that is responsiveness as illustrated in Figure 2B [16]. Localization refers to the initiation of repair activities locally at the site of damage without affecting the entirety of the structure. This could involve inclusion of different repair mechanisms for different positions or scales of the structure, for example, the healing mechanism at the surface and bulk could be different. Second concept is that of temporality, which means that the activation of the SH mechanism is temporary, operating only when needed. In the context of the SH materials mentioned, temporality is crucial because the healing action shall act only for a short duration, specifically between the time a crack occurs and the healing completion through cross-linking reactions. This is important because like in nature, healing does not happen spontaneously, and it tends to increase with time in most cases. This temporary nature ensures that the material retains its original properties and function after the healing process is finished, without ongoing changes or degradation over time. Mobility is another important concept for SH polymers as it directly affects the extent and speed of healing. Mobility in SH materials refers to the ability of certain components of the material or the polymeric chains to diffuse or move towards the site of damage. Mobility guides the localization and temporality rules, and if the system is restricted (e.g., due to crosslinking points), repairability would take longer or might not even occur [17]. Responsiveness, on the other hand, is the ability for the material to sense damage and environment and initiate the healing process effectively. Responsiveness enhances the system by enabling the polymer to react to environmental stimuli as well, such as temperature changes, pH shifts, or mechanical stress, which can trigger or accelerate the healing process [18]. This adaptive feature ensures that the SH mechanisms are not only present but also optimally active under conditions that typically accompany damage. Integrating responsivity into SH polymers promotes more robust and adaptable materials, capable of efficient self-repair across varying operational conditions.

## 2.3 | Healing Mechanisms

One of the simplest methods in which soft materials like polymers can heal is by physical approaches as outlined in Figure 2C. The physical SH process in polymers relies on several key mechanisms that enable damage repair and structural recovery. Interdiffusion involves the movement of polymer chains across damaged surfaces, allowing them to intertwine and restore the material's integrity [19]. In systems with phase-separated morphologies, the distinct phases can promote localized reflow or reorganization, facilitating autonomous healing [20]. Shape memory recovery leverages thermal or mechanical triggers to revert the polymer to its pre-damaged shape, closing cracks or fractures [21]. Additionally, melting interdiffusion occurs when elevated temperatures cause the polymer chains to melt and flow into damaged regions, effectively sealing the defects [22]. One of the initial approaches was the use of physicochemical interaction that involved integrating SH properties into polymer matrices, achieved through the incorporation of healing elements within microcapsules [23] or vascular networks [24]. This method, known as extrinsic healing, triggers the release of healing agents upon damage, effectively restoring material properties without external stimuli [25]. However, embedding

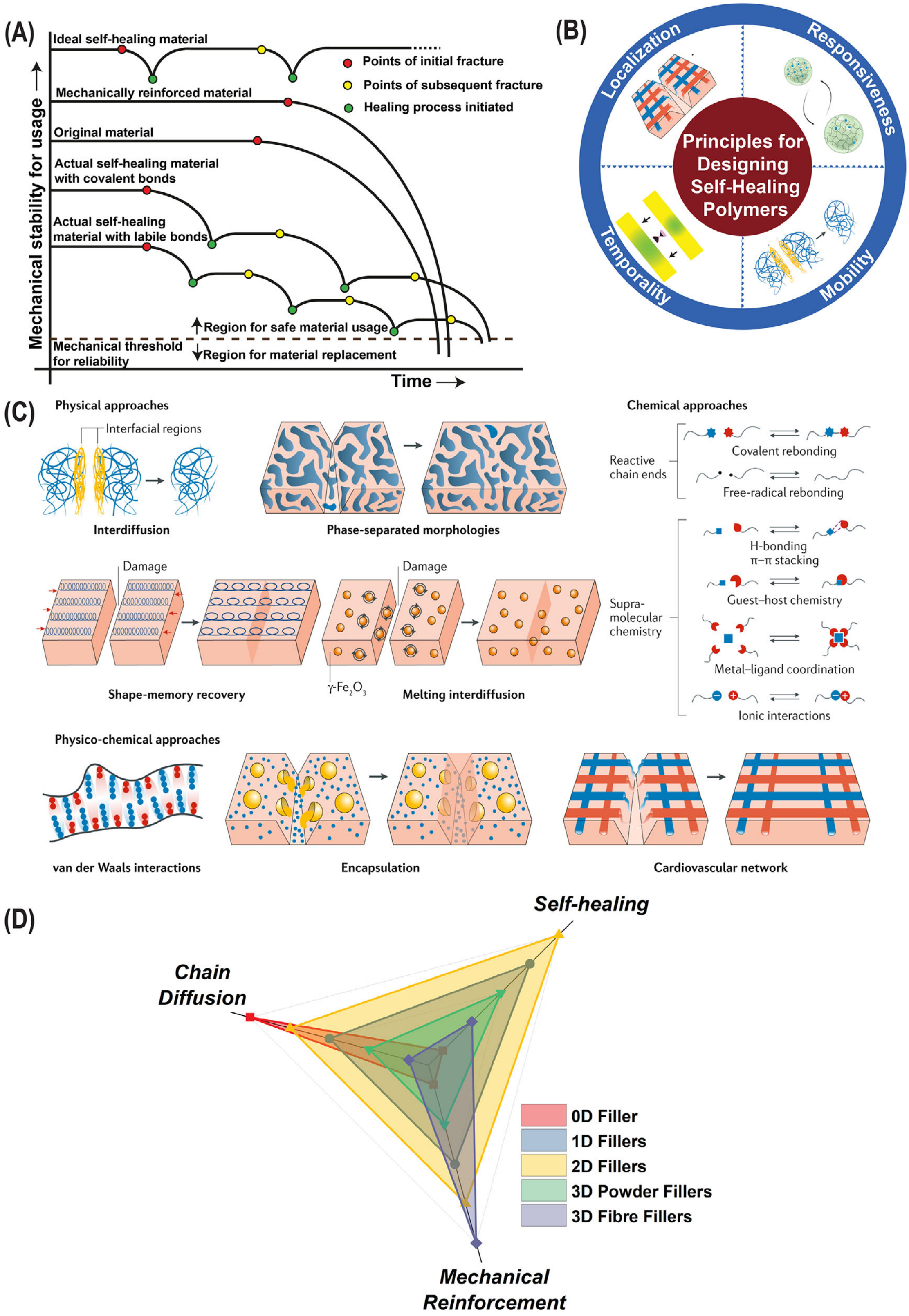

**FIGURE 2** | (A) Serviceable lifetime of materials with and without SH properties [28] (B) key principles for designing SH polymers. (C) Comparison of different fillers for SH polymers (D) mechanisms of healing in SH polymers [38]. *Source:* (A and B) Reproduced/Adapted from *Progress in Materials Science* [28] with permission from Elsevier. (C and D) Reproduced/Adapted from *Royal Society of Chemistry* [38] with permission from Springer Nature.

these healing components within the matrix often compromises mechanical strength and poses challenges in ensuring consistent healing due to limited healing agent availability [26]. Alternatively, intrinsic healing relies on chemical interactions with polymer chain diffusion and reversible bond formation inherent to polymer chains, allowing for repeated healing at the same site [27]. These are classified based on whether the reaction is based on reactive chain ends or uses supramolecular chemistry. Various reversible chemical bonding mechanisms such as DA bonding [28], metal–ligand coordination [29], disulfide bonding [30], diselenide bonding [31], acylhydrazone coupling [32, 33], Schiff-base [34], Michael addition [35], and urea bonding [36], among others, enable this process.

In multifunctional composites, the recovery of mechanical strength and functional properties often follows distinct mechanisms, which may not always align. Mechanical recovery typically involves the restoration of the material's structural integrity through processes such as chain entanglement, hydrogen bonding, or covalent bond reformation. This results in the partial or full restoration of properties such as tensile strength, fracture toughness, and impact resistance. In contrast, functionality recovery focuses on reinstating non-mechanical properties—such as electrical conductivity, barrier effectiveness, or thermal stability—which rely on the reformation of specific microstructures or conductive pathways. For example, in a recent study by Menon et al., joining the cleaved pieces of the composite nearly restored the electrically conductive pathways, and the electromagnetic interference (EMI) shielding capability remained practically unaltered after healing [37]. However, the recovery of mechanical properties was less than 70%, highlighting the disparity between the two recovery mechanisms. This demonstrates the importance of understanding the mechanism of functional healing in multifunctional composites, as functional properties may be the primary application of the material, making their complete recovery particularly desirable.

## 2.4 | Filler Type

Engineering applications often demand high-performance materials capable of withstanding harsh operational conditions. In many cases, polymers alone cannot fulfill these stringent requirements, necessitating their reinforcement with fillers that enhance mechanical, thermal, and chemical properties [39]. However, in the case of SH polymers, the choice of filler is particularly critical, as it must strike a balance between reinforcing the polymer matrix and maintaining a conducive environment for segmental chain motion during the healing process [14].

Traditionally, fillers hinder polymer chain mobility by reducing the free volume available for movement, thereby negatively affecting the healing process. This challenge, however, can be addressed through alternative strategies wherein the filler actively participates in the SH mechanism rather than merely serving as a structural reinforcement. For instance, fillers with high electrical or thermal conductivity can be leveraged to form percolation networks within the polymer matrix, enabling efficient distribution of external stimuli (e.g., heat or electrical current) to localized damage sites, thereby enhancing the healing process [40]. Additionally, the alignment of fillers within the matrix can facilitate the formation of microchannels, promoting unidirectional polymer chain diffusion and improving healing efficiency [41]. Surface energy modification is another approach, where fillers influence interfacial interactions and drive polymer chain migration, promoting the reformation of bonds at the damage site [42]. Furthermore, functionalizing filler surfaces with reactive moieties can enhance SH by increasing the number of potential reconnection sites, while simultaneously improving interfacial adhesion between the filler and the polymer matrix, resulting in a stronger composite [43].

Beyond the filler's interaction with the polymer, its morphology plays a critical role in determining the final composite properties. Among the most common fillers for polymers, five different morphologies stand out due to distinctive properties, and these also help understand the spectrum of fillers and their impact on properties of the composite. Figure 2D presents a comparative radar plot illustrating the influence of various filler morphologies like 0D, 1D, 2D, and 3D fillers (including powders and fibers) on key performance attributes relevant to SH polymer composites, namely, chain diffusion, SH efficiency, and mechanical reinforcement. The data highlight the inherent trade-offs among these properties. Although 0D fillers promote chain diffusion, which is crucial for SH, their limited mechanical reinforcement constrains their practical application [44]. In contrast, 3D fiber fillers significantly enhance mechanical properties but impede chain mobility, thereby diminishing SH efficiency [45]. 1D and 3D powder fillers exhibit intermediate characteristics, offering a compromise between reinforcement and SH potential [46]. However, 2D fillers emerge as the most effective solution in achieving a balance between mechanical reinforcement and SH, while maintaining moderate chain diffusion [11]. This optimal combination is attributed to their high aspect ratio, which facilitates stress dissipation and network connectivity without excessively hindering polymer chain dynamics [47]. This makes 2D materials particularly promising for the design of intrinsically SH polymer composites, offering a synergistic enhancement of structural integrity and reparability. Each of these materials offers unique advantages, including exceptional mechanical reinforcement and enhanced barrier properties due to their layered structure and high aspect ratio. Their key properties and selected applications are summarized in Figure 3 [39].

Polymer composites incorporating 2D fillers have gained significant attention due to the exceptional properties imparted by the unique flake-like shape and high aspect ratio of these materials [48]. The use of 2D materials as fillers enhances the composites' strength-to-weight ratio, mechanical strength, flexibility, optical transparency, and wear resistance. Additionally, they impart multifunctional properties, including electrical and thermal conductivity, flame retardancy, sensing capabilities, and environmental protection [49]. The distinctive morphology of 2D materials allows them to efficiently occupy the matrix bulk and reinforce it through improved interfacial contact due to their high surface area. Moreover, they can form percolation paths that enhance properties like electrical conductivity, even at low weight loadings.

For SH polymers, 2D nanofillers can participate in the active healing process (filler-dependent healing) by forming reversible bonds with reactive sites in the matrix, thanks to surface

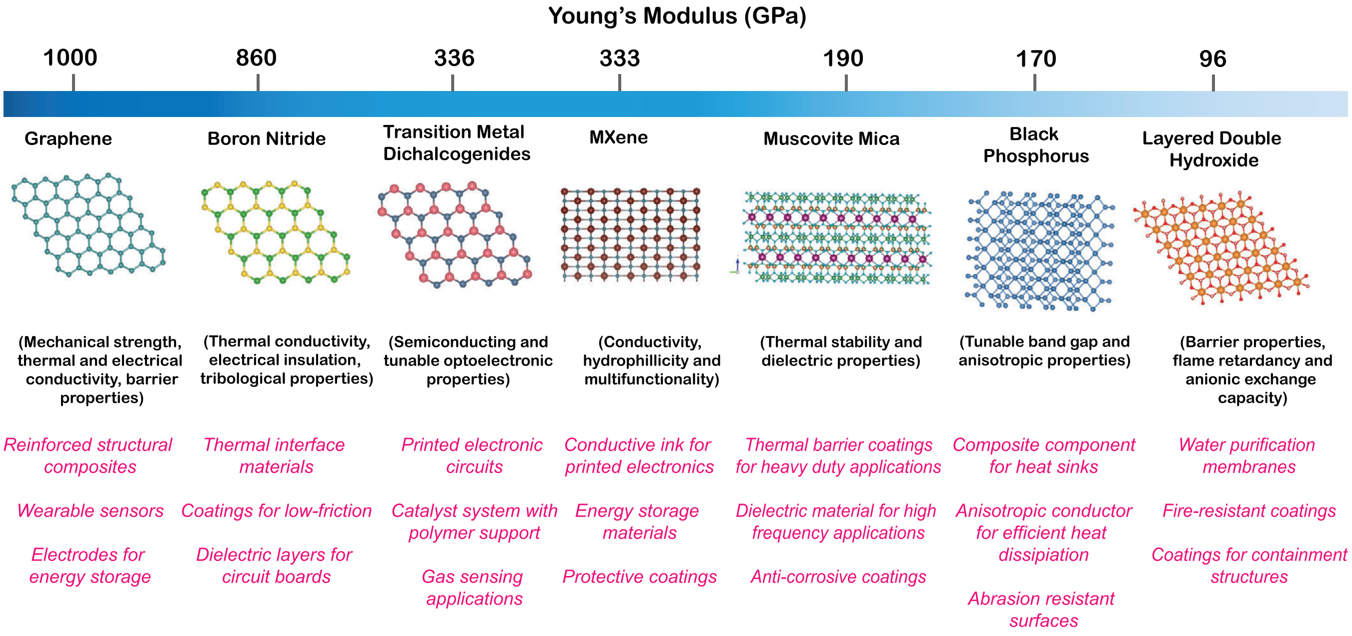

**FIGURE 3** | Different 2D materials with their properties and potential applications.

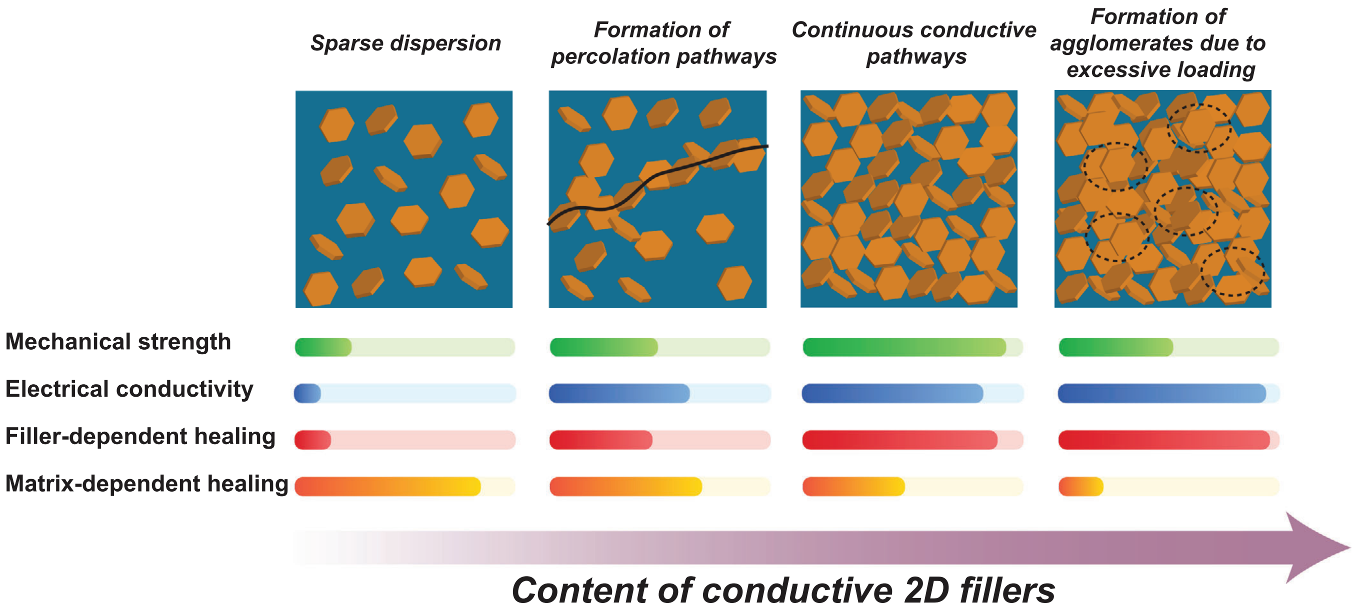

**FIGURE 4** | Effect of 2D nanofiller loading on the mechanical, electrical, filler-dependent healing, and matrix-dependent healing properties.

functional groups [43]. Alternatively, these nanofillers can convert applied energy stimuli into forms usable by the composite for healing [50]. In some cases, these 2D materials act as passive fillers (matrix-dependent healing), where they do not directly participate in the healing chemistry but instead reinforce the matrix, with the matrix itself driving the healing process [51]. The effectiveness of these roles depends on factors such as the type of 2D material used, its surface properties, dispersion within the host matrix, and the nanofiller's weight loading. The amount of 2D material loaded into the composite significantly impacts its final properties. For instance, as shown in Figure 4, increasing filler loading initially boosts mechanical strength until the point where the nanosheets are well dispersed in the matrix. Beyond this point, further loading leads to agglomeration (circled), making the composite more prone to failure. Electrical conductivity, on the other hand, directly correlates with the filler quantity where formation of percolating pathways is the key. Evaluating healing properties is more complex, as healing can be defined in several ways (as discussed later in the article). When based on mechanical properties, healing can be filler-dependent, directly tied to the amount of 2D filler, or matrix-dependent, where it inversely correlates with the filler quantity.

## 3 | SH Polymer Composites Based On Graphene

Since its discovery in 2004, graphene has captured significant attention in the scientific community due to its remarkable mechanical strength, outstanding electrical and thermal conductivity, and other desirable properties [52]. Graphene, which is a two-dimensional carbon allotrope, consists of a single layer of $sp^2$-bonded carbon atoms arranged in a hexagonal lattice structure, resulting in an ultra-thin, lightweight, and robust material with large specific surface area and energy-absorbing capabilities [53, 54]. Its properties have led to extensive research exploring its applications in fields such as energy [55], desalination [56], composites [57], coatings [58], sensors [59], membranes [60], and electronics [61]. Recently, it was also found that graphene-based materials like graphene oxide (GO) can be synthesized by safer processes, where even direct inhalation by humans did not have any adverse effects on cardiorespiratory responses [62]. The versatility and immense potential of graphene have made it a subject of great interest and investigation in the pursuit of developing advanced materials and technologies [63].

The crystalline nature of graphene allows for the attachment of other atoms to its surface, leading to the formation of diverse graphene variants, each possessing unique properties and applications. These modifications can alter the mechanical, electronic, optical, and chemical properties of graphene, expanding its potential applications in various fields [64]. The ability to tailor graphene's properties through surface functionalization opens up new avenues for research and development of graphene-based materials with customized characteristics to meet specific application requirements. The performance of graphene derivatives, along with their tunability to match the host matrix, motivates ongoing research in the field of graphene–polymer composites [65]. Various graphene-based additives like pristine graphene, GO, reduced GO (rGO), and even functionalized graphene have been used in the past to synthesize polymer-based SH nanocomposites [66]. Moreover, the preparation and functionalization methods for graphene have reached a mature stage, including mechanical exfoliation, liquid phase exfoliation, high shear exfoliation, and plasma chemistry that enable production of economically viable graphene at commercial scale [67]. Graphene-based SH polymer composites have found diverse applications in various fields, including biological applications, sensors, coatings, supercapacitors, flexible electronic devices, artificial skin, self-cleaning surfaces, adsorbents, biomimetic materials, and actuators [66].

In general, polymeric materials are prone to failure and loss of sustainability, safety, and durability when they experience fractures or damage during service. Consequently, researchers have focused on finding solutions for repairing these materials. Although the concept of completely autonomous self-repairing materials is ideal, it often results in lengthy repair times [68]. Therefore, external assistance is typically required to restore the original performance of damaged materials. External assistance or stimulus can come in various forms, including heat, light, mechanical force, electromagnetic waves, moisture, pH, and electricity [66]. Graphene as an additive can accentuate the effect of stimulus due to its superior properties, and this enables efficient utilization of the applied energy within the healing system. Graphene derivatives like any other 2D material can exist in three different forms within a polymeric matrix: (a) being attached to the matrix through intermolecular forces [69], (b) being attached to the matrix via hydrogen bonds [70] or host–guest interactions [71], and (c) being crosslinked with the polymer matrix through reversible chemical bonds [72]. In the following section, graphene-based SH materials have been classified according to their mode of healing, and a comprehensive explanation of the SH pathways employed is provided.

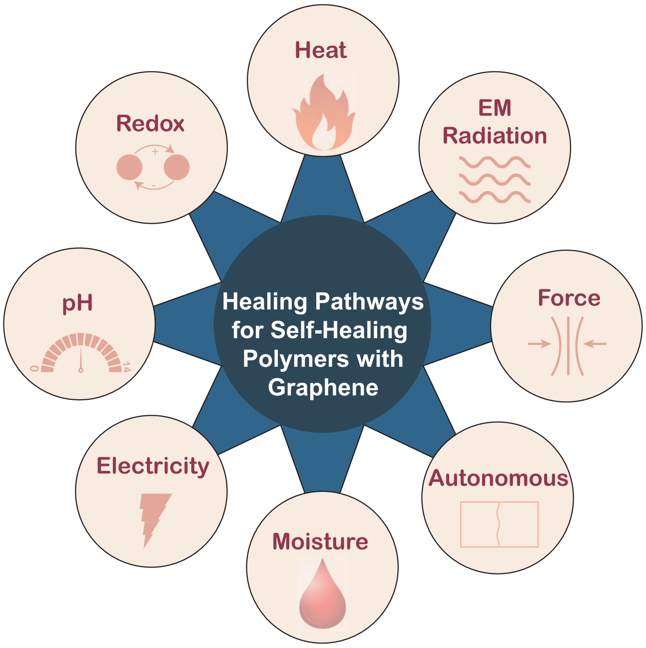

**FIGURE 5** | Healing pathways for graphene-based SH polymer composite.

In graphene-based composites, SH can occur through various methods when triggered by external energy or stimuli, also known as non-autonomous healing. These stimuli include heating, light radiation, microwave energy, chemical change, and solvents as shown in Figure 5. Sections below are classified on the basis of stimuli needed to heal graphene-based SH polymer composites.

### 3.1 | Heat

The application of heat is widely employed as a stimulus to initiate SH in polymers. This is primarily due to the increased movement of polymer chains as they approach the glass transition temperature [73]. The elevated temperature promotes the formation of reversible bonds, allowing for temperature-dependent reversibility in healing mechanisms such as the DA reaction to take place [28]. Due to the excellent thermal conductivity of graphene, polymers can undergo the SH processes and regain their structural integrity through the effective utilization of heat [74]. Numerous studies have reported successful healing of the graphene-based polymer nanocomposite (PNC) by employing heat as the source of stimulus and show that graphene improves the thermal-healing behavior of polymers [75–85]. For instance, a conductive flexible sensor with repeatable SH properties was synthesized by Tu et al. using functionalized GO and waterborne polyurethane (WPU), which could heal effectively at 130°C [86]. Similarly, epoxy resins loaded with bismaleimide-modified graphene nanoplatelets (GNP) could be healed with 87% efficiency at the same temperature [72]. Most of the bonding chemistries employed for SH of polymers are sensitive to temperature and give increased healing efficiency when the temperature increases. For example, DA-based systems have reversible covalent bonds that connect and disconnect at a temperature based on the type of diene and dienophile [28]. On the other hand, heat-induced segmental movement of the polymeric chains causes chain repositioning within the matrix, thereby enabling the formation of new bonding sites among the polymer chains as well as with the fillers [66]. The temperature at which such interactions happen is determined by the thermodynamic requirement of the bonds to be formed and the thermal characteristics of the polymer.

### 3.2 | Electromagnetic Radiation

Alternative stimuli sources, such as indirect heating methods, offer certain advantages in terms of SH in polymeric compounds. This could directly benefit the structure by preventing material degradation that can occur due to direct heat exposure. Additionally, they offer a more convenient pathway for SH, catering to the specific requirements of the polymeric compounds where heating the materials, whereas application is difficult. Microwaves can be converted to heat in the medium, and this heat can be utilized to break and make the bonds. Many previous works have reported efficient healing using microwaves, but notably, Wong et al. produced covalently cross-linked polyurethane (PU)/rGO with excellent healing properties. This way, the utilization of microwaves in conjunction with graphene can facilitate heat generation, enabling the reversible alteration of DA bonds. This process promotes the wetting, movement, and intertwining of polymer segments, ultimately healing the composite [87]. Due to excellent photothermal properties of graphene, such composites can also be used to heal the graphene-based composite by converting the irradiated ultraviolet (UV)/visible light into heat. This triggered an interesting line of research where infrared (IR) [88–93], green light [94], or even direct sunlight [95] has been used to restore the mechanical properties of graphene-based PNCs. Some other works have reported multiple healing pathways like heat, IR light, and microwaves for same material, which also demonstrates the superiority of microwave healing over the other methods. Microwave healing can also result in healing efficiencies greater than 100% as they produce bulk heat when healing and tend to increase the degree of polymerization (number of sites for reversible bonding) in chemically bonded healing networks [96].

### 3.3 | Mechanical Force

In the case of most polymer composites where healing happens at room temperature without any other external energy, mechanical force is usually required to join the pieces together and initiate the rebonding process over the period of time. This force may be almost negligible and might only need a gentle push to the cleaved pieces, and it is, however, classified as healing through mechanical force pathway. This mostly involves polymeric hydrogels that are SH and self-adhesive in nature but need placement of cleaved pieces in close proximity to initiate formation of bonds. Wang et al. prepared an amine-terminated randomly branched oligomer network with thionyl chloride-modified GO nanosheets that could heal at room temperature by manually joining the pieces together with a healing efficiency of almost 90% based on a hydrogen bond network [97]. Exploiting the supramolecular interactions between polymer–polymer and polymer–graphene, room-temperature healable copolymer frameworks of 2-[[(butylamino)carbonyl]oxy]ethyl ester (PBA)–polydimethylsiloxane (PDMS) were also prepared with a healing efficiency of almost 100% [98]. This work also suggests possible mechanisms in Figure 6 by which broken 2D nanosheets at the interface can heal after the healing of the macrostructure is completed. In Figure 6A, it can be seen that the healing of the composite basically constitutes polymerization, which is the healing of macromolecular backbone and healing of graphene framework. Figure 6B shows macroscopic healing process where chain diffusion is shown to be diverted towards the crack interface, and Figure 6C shows the mechanisms of skeleton adhesion, polymer adhesion, and skeleton hook, which can occur in healing of the graphene framework and restore the thermal transport pathway. Another interesting observation is that mechanical strength and thermal conductivity recovery follow distinct pathways, with mechanical strength exhibiting significantly higher recovery. This is because mechanical recovery is aided by physical chain entanglement, which allows the polymer matrix to regain its structural integrity. In contrast, the recovery of thermal conductivity relies on the restoration of conductive pathways, which requires the healing of broken graphene nanosheets. However, because this nanosheet restoration does not occur within the composite, thermal conductivity experiences a permanent loss with each healing cycle.

### 3.4 | Autonomous Healing

Among the various healing mechanisms, autonomous healing is considered the most desirable because it does not require any external intervention to initiate or propagate the healing effect. No additional forms of energy, such as heat or electricity, are necessary, and most of such structures function by instigating the healing process through internal mechanisms usually triggered by perceiving the damage. Extrinsic types of healing systems based on microcapsules have been well documented [99–102]. However, in terms of intrinsic healing, D'Elia et al. presented a prominent study where a supramolecular polymer was synthesized with PDMS as a base matrix and integrated into a graphene ultralight network to create durable and electrically conductive composites [103]. These composites exhibit pressure-sensing capabilities and possess the remarkable ability to fully restore their properties even after multiple instances of damage, without requiring any external stimuli. The autonomous healing in this work is driven by the natural formation of dynamic bonds between the boron and oxygen atoms on the silicone backbone, along with the formation of hydrogen bonds with the polymer chains.

### 3.5 | Other Pathways

Apart from the traditional stimulus sources, there have been advancements in the development of alternative healing pathways for graphene-based SH polymers. For example, a highly conductive polyelectrolyte film was prepared using polyacrylic

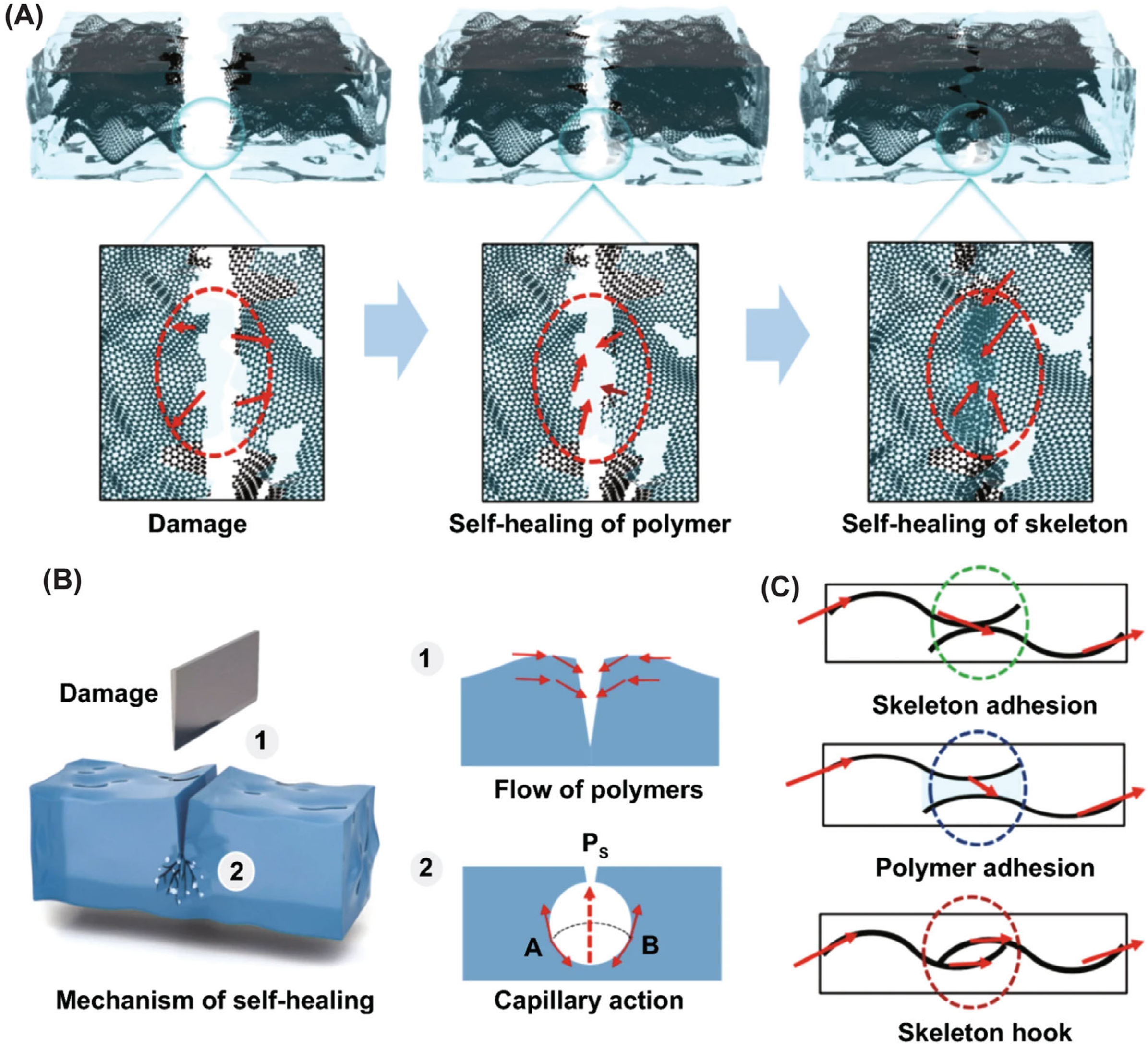

**FIGURE 6** | (A) Schematic of the SH process of PBA–PDMS/graphene (B) SH mechanism of macroscopic damage to materials. (C) SH mechanism of PBA–PDMS/graphene for thermal conductivity [98]. *Source:* Reproduced/Adapted from *Nano-Micro Letters* [98] with permission from Springer Nature.

acid (PAA), branched polyethylenimine (PEI), and graphene through a layer-by-layer self-assembly method, which could heal efficiently in the presence of high humidity [104]. Several other works have also reported SH triggered by moisture/water, where $H_2O$ molecules help in the reconstruction of hydrogen bonds between cleaved chain ends [105–107].

Electricity has quite often been employed to impart indirect heating to conductive materials by using the principles of resistive heating [108]. As can be imagined, this healing pathway is only limited to composites with measurable electrical conductivity. The addition of graphene/rGO to the polymer matrix with proper dispersion has the ability to make the polymer electrically conductive; however, highly oxidized/functionalized derivatives do not offer this advantage due to low inherent electrical conductivity [109]. Huang et al. used a similar technique and fabricated a novel SH composite with few-layer graphene and PU that showed repeatable healing behavior by electricity with almost 98% healing efficiency [89]. This comes in conjunction with a study published by Liu et al. where the group has shown a solder-free technique to “weld” macroscopic graphene sheets by electrical joule heating. It appears highly feasible to achieve defect-free covalent cross-linking between graphene layers using Joule heating at the cleavage sites of graphene nanosheets that are exposed at the interface of the split composite pieces [110].

Furthermore, Cong et al. prepared a dual network poly(acryloyl-6-aminocaproic acid) and GO-based mechanically strong hydrogels with $Ca^+$ ions reinforcing the structure with coordination bonds [70]. With a robust hydrogen bonding network between the polymer chains and the GO edges, it showed pH-triggered SH behavior in acidic environments at $pH \leq 3$ and cleaving properties at $pH \geq 7$. Triggering a healing response to the chemical environment is rarely observed, and such systems have tremendous potential for biological scaffolds and drug delivery applications where assembly and disintegration on demand are very desirable.

The utilization of host–guest interactions for SH polymers is particularly relevant when addressing the main issue associated with nanofillers such as graphene, which can impede the motion of polymeric chains within the matrix and hinder the healing process. In a recent work by Xiang et al., self-healable composite films comprising PAA and branched PEI chemically functionalized with ferrocene and $\beta$-cyclodextrin ($\beta$-CD) modified rGO were synthesized using a layer-by-layer approach [111]. When the films are cleaved, healing can be initiated by exposing the cleaved surfaces to an oxidative environment ($H_2O_2$) where healing occurs by breaking host–guest interactions, followed by exposure to a reducing environment (aqueous glutathione solution) that helps to restore the mechanical strength of the film. Some other healing mechanisms, such as bionic healing [112] and catalytic crosslinking [113, 114], have also shown potential for graphene-based SH polymers; however, further studies are required to understand how such mechanisms can be translated into applications.

## 4 | SH Polymer Composites With 2D Materials Beyond Graphene

Nobel laureate Richard P. Feynman once posed significant questions during his lecture titled “There’s plenty of room at the bottom,” asking, “What could we do with layered structures with precisely arranged layers? What would be the properties of materials if we could manipulate atoms as desired?” [115]. In this era, we are witnessing the realization of Feynman’s vision with the discovery of novel 2D materials and the development of sophisticated techniques to modify them at the atomic level [49, 116, 117].

Through the manipulation of layers, the opportunity arises to design materials with exceptional physicochemical characteristics. Upon examining a diverse range of investigated van der Waals (vdW) solids and their distinctive sets of properties, it becomes evident that these materials hold immense promise [118]. Many of these 2D materials have shown exceptional potential in composites and facilitated damage repair by various SH mechanisms [119, 120]. Outlined below are key 2D materials that have been utilized in the development of SH polymer composites. The subsequent discussion includes their distinctive properties, methods of preparation, recent advancements concerning SH polymer composites, and their unique contribution to the composites’ properties.

### 4.1 | Transition Metal Dichalcogenides (TMDs)

TMDs consist of individual layers composed of a transition metal atom sandwiched between two chalcogen layers. These layers can be stacked in three different crystal structures: a trigonal prismatic phase (2H), a rhombohedral phase (3R), and an octahedral phase (1T), with the last one exhibiting metallic behavior [121]. The transition between these phases can occur through the gliding of atomic layers [122]. In the 2H phase, which is the most commonly studied crystalline form of TMDs, the layers exhibit a distinctive ABAB-layer stacking pattern, where the sulfur (S) atoms of one layer align with and eclipse the transition metal atoms of the adjacent layer [123]. TMDs have been successfully employed for energy storage [124], bio sensors [125], anticorrosive coatings [126], piezoelectric sensors [127], and photonic devices [128]; however, they are better known for their excellent semiconducting [129–131] and lubricating properties [132, 133]. Regardless of their inherent superior properties, TMDs have been vastly researched due to the possibility and ease of fine-tuning their properties. Bulk 2H crystals of such TMDs behave as indirect band-gap semiconductors (with band gap of 1–1.29 eV for $MoS_2$, 1.3–1.4 eV for $WS_2$, 1.1 eV for $MoSe_2$, and 1.2 eV for $WSe_2$) [134]. However, when the monolayer counterparts of these crystals are observed, they behave as direct band-gap semiconductors (with band-gap 1.8–1.9 eV for $MoS_2$, 1.8–2.1 eV for $WS_2$, 1.5–1.6 eV for $MoSe_2$, and 1.6–1.7 eV for $WSe_2$) [135]. The presence of direct band-gap and high light absorption, along with their electron mobility equivalent to silicon, means TMDs can be a crucial class of materials for highly efficient transistors and optoelectronic devices [136]. When it comes to tribological properties, 2D $MoS_2$ has proven to be even better than graphene for lubrication under load. Similar results have been obtained when 2D $MoS_2$ has been used either as a dry lubricant, lubricant additive, or coating [137, 138].

Synthesis of TMDs has been studied for many decades. $MoS_2$ in some of the early works was synthesized by reduction of $MoS_3$ with hydrogen [139, 140] or by solution processing techniques usually occurring at high temperatures [141]. The technique for synthesis is highly dependent on the end use case and potential application. For example, the chemical vapor deposition (CVD) technique is highly suitable for large-area electronics, whereas liquid exfoliation can help in bulk production of nanosheets for lubricants and composites. Various techniques are employed for the synthesis of 2D TMDs as summarized in Figure 7 [142]. These methods encompass powder vaporization, chemical vapor transport, metal transformation, chemical exfoliation, molecular beam epitaxy, CVD, spray pyrolysis, pulsed laser deposition, and electrochemical synthesis.

Two-dimensional forms of these TMDs have proven to be very useful in reinforcing the structure of polymers and assisting in improving the mechanical and SH properties of the nanocomposite. Even though some TMDs are susceptible to oxidation when exposed to light and air [143], few of the advantages brought into the polymer matrix by the addition of TMDs include increased chemical resistance, improved flame retardancy, enhanced barrier properties, biocompatibility, improved electrical conductivity, and improved wear resistance [144].

Many works previously have utilized excellent photothermal properties of TMDs to provide external stimulus to the hosting polymeric matrix and facilitate healing under light [145, 146]. Due to the highly layered structure and high intrinsic absorption band gap of TMDs like $MoS_2$, near infrared (NIR) light can be easily absorbed. This absorption causes local surface plasmon resonance (SPR) effect by lattice vibration or electronic oscillation where the absorbance of photon triggers recurring electron oscillations [147]. When $MoS_2$ is bombarded with NIR light, valence band electrons move from the ground state to the excited state, and when these electrons return to the ground state, they release the absorbed energy in the form of heat, causing localized hot-carriers [148] and dissipating this heat to the host polymer matrix. This heat can trigger the stimuli-based healing response in the composite by debonding/bonding along with enabling chain mobility by closing the gap to the glass

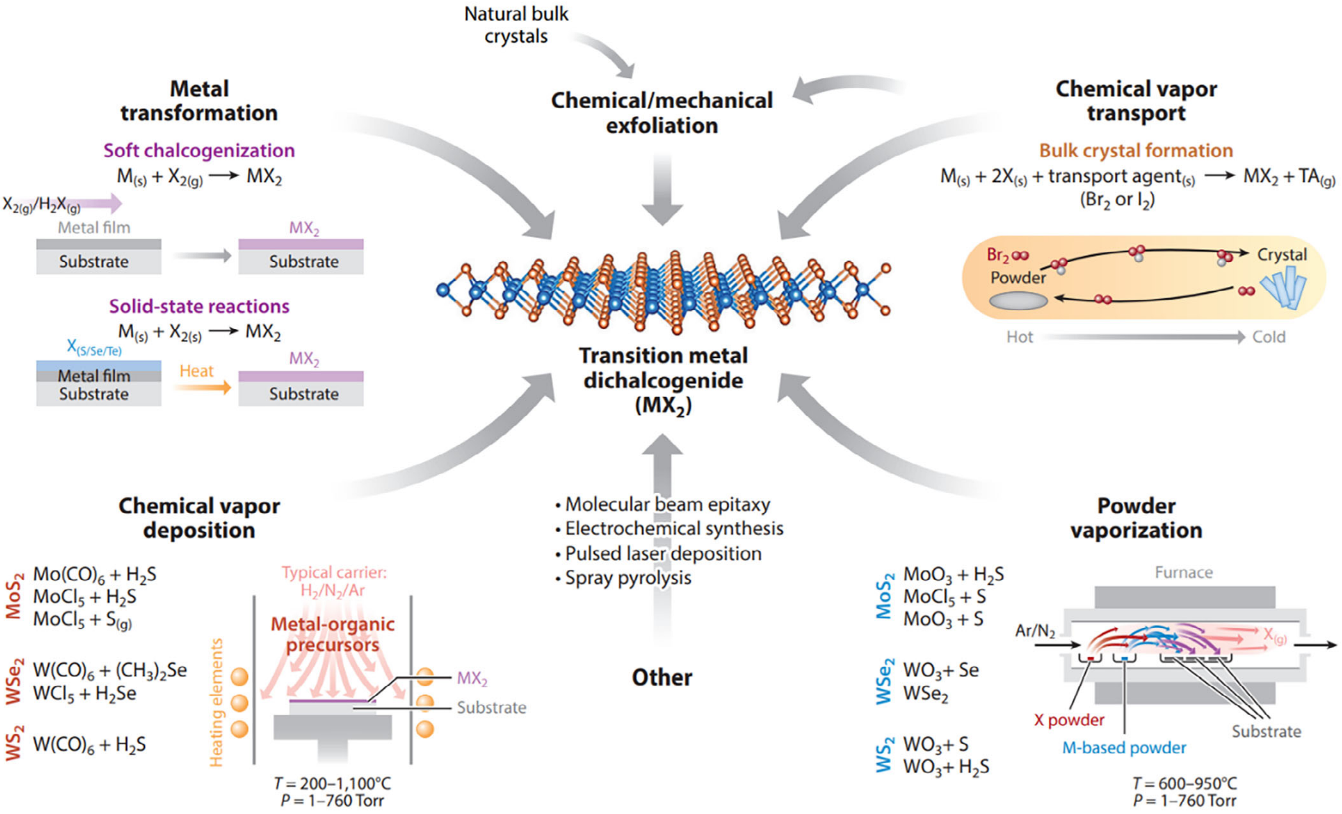

**FIGURE 7** | Summary of production techniques for 2D TMDs [142]. *Source:* Reproduced/Adapted from *Annual Review of Materials Research* [142] with permission from American Chemical Society.

transition temperature. This effect was exploited by Zheng et al. where IR-driven photothermal effect was used to induce SH in $MoS_2$/polyvinyl alcohol (PVA) hydrogels based on hydrogen bonding, resulting in a 612% increase in tensile strength and almost 90% healing efficiency. In this work, it is also shown that temperature increase in flower-like $MoS_2$ is much higher than sheet-like $MoS_2$ structure when exposed to NIR, resulting in rapid and better healing characteristics, suggesting an effect of morphology on photothermal properties of the nanofiller. Furthermore, it can also be observed that flower-like $MoS_2$ results in a stronger nanocomposite as the 3D structure of the nanomaterials reinforces the material by 3D steric hindrance as compared to 2D nanosheets. This highlights the importance of nanofiller morphology in designing nanocomposites with SH abilities, which is often ignored [50]. In a similar work by Yang et al., even higher healing efficiency was obtained by using chemically exfoliated $MoS_2$ nanosheets and PVA with similar bonding mechanisms that could also be due to the contribution from polymer matrix as PVA with higher molecular weight was used [149].

When looking into potential applications of such self-healable TMD/polymer composites, Menon et al. used dopamine-functionalized PU combined with mussel-inspired flower-like magnetic $MoS_2$ structures and used the nanocomposite as an EMI shielding material. The healing mechanism employed was based on the formation of reversible dynamic coordination bonds between hydroxyl functional groups introduced by dopamine and $Fe^{3+}$ ions from magnetic $MoS_2$. Nanocomposite shows a shielding capability of −36.6 dB with up to 96% absorption. When this material was healed by shape memory assisted self-healing (SMASH), it showed almost 56% recovery in tensile strength [150]. An ultrathin, lightweight, and thermally healable EMI suppressor was also developed by the same group using $MoS_2$ as electric dipoles and forming a healable network using reversible metathesis reaction [151]. In another work, Xu et al. deployed a self-healable $MoS_2$-polyethylene glycol (PEG)-based hydrogel for sensing applications, showing fast healing characteristics (90 s) at an ultra-low nanosheet loading of 0.04 wt.%. This sensor can be used for both contact and non-contact sensing properties due to its light-sensitive and deformation-dependent conductivity [152].

In a recent remarkable discovery, the biocompatibility of bio-derived polymeric materials and excellent photothermal conversion efficiency of TMDs have been used in conjunction to synthesize SH polysaccharide hydrogels for photothermal-photodynamic therapy to treat breast cancer. Such bio-based nanocomposite hydrogels can heal autonomously and facilitate intratumoral injection by delivering therapeutic molecules on site, rather than unwanted dilution caused by intravenous injection. To increase the biocompatibility of $MoS_2$, nanosheets were wrapped in bovine serum albumin before blending with the hydrogel. This solves a major problem where photothermal agents and photosensitizers interfere with each other and cannot work in synergy during the treatment [153].

Inspired by biological cartilage, Wang et al. produced tannic acid-modified $WS_2$ nanosheets, incorporated within the healable PU network to produce a high-density network of noncovalent bonds, producing strong interaction between the nanosheets and polymer matrix at the interface. The nanocomposite network shows excellent tensile strength, toughness almost 10 times higher than metallic aluminum, and excellent healing efficiency (80%–100%). Impressive mechanical properties were obtained without compromising the healing efficiency, which is quite rare in healable networks with non-covalent bonding. This ultra-strong and highly flexible nanocomposite was further tested for its use in actuation devices [154].

Most of the works discussed till now focus on ex situ blending of polymers and nanosheets where $MoS_2$ rarely participates in the construction of the host matrix, and it rather acts as a nanofiller and reinforces the matrix for within. However, in a recent work by Lee et al., $MoS_2$ was employed to promote the radical formation and thereby polymerize polyacrylamide (PAM) under mild conditions without any external stimuli or crosslinking agent. Here, $MoS_2$ also undergoes a polymerization-induced phase transformation from 2H to 1T phase. However, when $MoS_2$ concentrations higher than 0.2% were used, the hydrogel composites were no longer flexible. Much of the tensile strength of the hydrogel could be recovered when cut pieces were joined together and heated at 70°C [155].

Recent progress in the functionalization of TMDs has opened avenues towards the synthesis of self-healable TMD/polymer composites with various bonding mechanisms. For example, Perez and group attached maleimide-based moieties on $MoS_2$ and $WS_2$ nanosheets by mild covalent functionalization that can be further used in conjunction with furan and maleimide functionalized monomers to yield a healable network based on DA chemistry [150]. Other TMDs such as $MoSe_2$ and $WSe_2$ and even pnictogen chalcogenides such as $Sb_2S_3$ and $Bi_2S_3$ have previously been functionalized using aryl diazonium salts with almost eight hydrogen bond acceptor counts on each molecule [151]. Having higher hydrogen bond donor/acceptor sites makes it easy to facilitate the formation of a crosslinked H-bond network with the host matrix. Most functionalization techniques involving TMDs work on defect engineering; however, Voiry et al. developed a functionalization technique based on electron transfer between TMDs ($MoS_2$, $WS_2$, and $MoSe_2$) and an organohalide [152]. Such novel methods to functionalize relatively inert TMD nanosheets need to be developed to impart desirable functional groups that have the potential to participate in repeated bonding/debonding reactions. In most cases yet, $MoS_2$ has been used as a model TMD to show the modification and application in SH polymers; however, other TMDs are not widely explored.

### 4.2 | hBN

2D hBN nanosheets, characterized by $sp^2$ hybridization, are also commonly referred to as "white graphene," and they represent the insulating structures in a 2D format [156]. They share a structural likeness to graphene, possessing a honeycomb arrangement where the sublattices are filled with boron and nitrogen atoms in alternating patterns. The hexagonal crystal structure of hBN is defined by crystallographic dimensions $a = 0.250$ nm and $c = 0.666$ nm, and the gap between layers is 0.333 nm. hBN is an electrical insulator and thermal conductor that possesses high mechanical strength, high surface area, high band gap, high resistance to oxidation, optical transparency, and lubricating properties [157, 158]. It can be synthesized by various methods as shown in Figure 8.

Although both hBN and graphene nanomaterials exhibit weak intermolecular vdW forces and $\pi$–$\pi$ interactions with host matrices like polymers, the partial charge separation caused by some degree of electron transfer inherent to the B–N bonding offers a potential advantage in terms of molecular interactions with polymeric materials [159]. hBN differs distinctly from the hexagonal graphitic structure of graphene due to the distinctive nature of its heteroatom bonding. This distinction arises from several factors, mainly the partially ionic character of the B–N bond, the involvement of a p-bond where an empty p-orbital of boron interacts with a filled orbital of nitrogen, and the specific positioning of an electron closer to nitrogen in the s-bond [160]. This differentiation in electron distribution results in the limited delocalization of nitrogen's electrons, and the presence of ionic character within the bond disrupts the symmetrical electronic state. This also prompts the emergence of a band gap in hBN sheets [161].

According to molecular dynamics simulations, the interplay between polymers and hBN nanomaterials has the potential to offer superior bonding efficacy in contrast to carbonaceous nanofillers [162]. This enhanced interaction is attributed to the inherent polarization in B–N bonds, the strong bonding capabilities of B and N atoms, as well as vdW forces and Coulombic interactions between the hBN nanomaterials and the host matrix [163]. Consequently, by skillfully designing and synthesizing nanocomposites, there arises a promising opportunity for optimizing and altering the chemical interactions on a molecular level.

Owing to these advantageous attributes, the exploration of hBN/polymer composites has primarily centered on harnessing hBN's capabilities for enhancing thermal conductivity while maintaining electrical insulation within the composites [164]. In more recent times, an increased amount of research has been dedicated to unraveling the intricate mechanisms responsible for the mechanical reinforcement effects of hBN when integrated into polymer matrices [165]. Extensive efforts have been directed towards comprehending the interplay between hBN's unique properties and the structural dynamics of the polymer matrix, aiming to uncover novel avenues. When it comes to mechanically reinforcing polymer composites, hBN has emerged as a reliable reinforcement across a range of polymers such as PVA [41], polyvinyl chloride (PVC) [166], polyethylene (PE) [167], polytetrafluoroethylene (PTFE) [168], polyvinylidene difluoride (PVDF) [169], polycarbonate (PC) [170], and epoxy [171].

The infusion of SH attributes into hBN-infused polymer composites by solution mixing [172] or in situ polymerization [173] has garnered significant attention in recent times. This forward-looking trajectory leverages the exceptional mechanical and thermal transfer attributes of hBN in conjunction with the innate self-repair capabilities of polymers. In one of the earliest works by Lafont et al., temperature-responsive SH composites based on a reversible disulfide bonding mechanism, featuring 3D hBN and graphite particles within a polysulfide-based thermoset matrix, were fabricated for enhanced thermal conductivity [174].

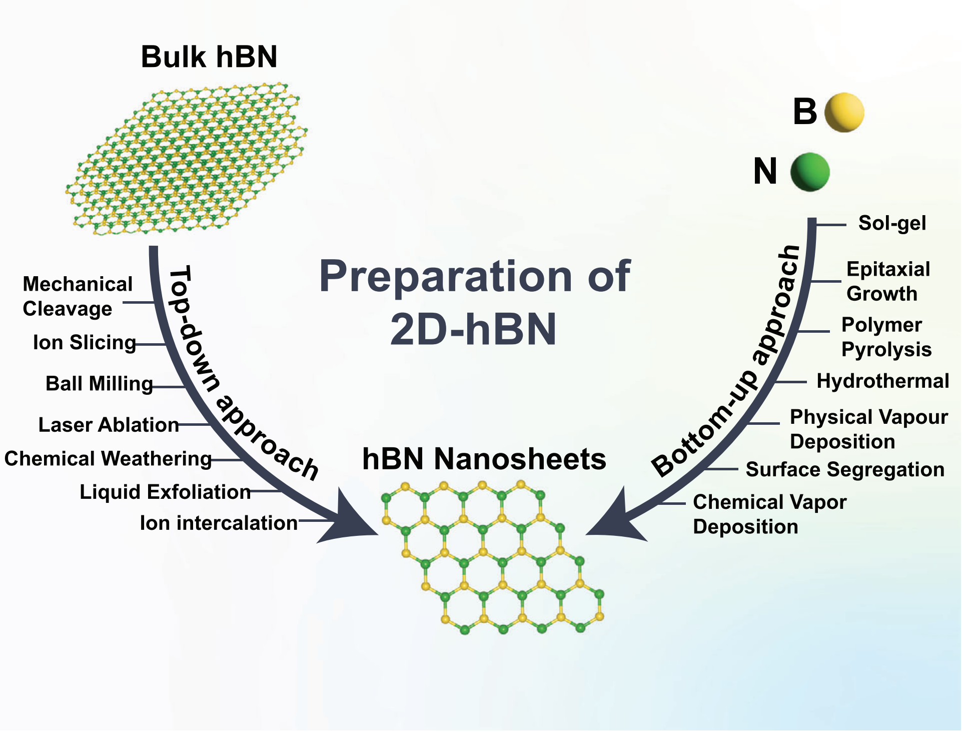

**FIGURE 8** | Summary of methods to synthesize 2D hBN.

These composites could recover their cohesive and adhesive properties through thermal activation, with full adhesive strength restoration at a mild 65°C healing temperature over multiple cycles. Cohesion recovery varies (20%–100%) based on filler type, loading, and matrix. Thermal conductivity increases with filler content, reaching 1 and 2 W/m K for boron nitride and graphite composites, respectively. These findings demonstrate the design potential for multifunctional materials with distinct mechanical SH capabilities using thermal healing principles.

Interfacial adhesion stands as a pivotal consideration within the domain of SH nanocomposites [175]. Often, the introduction of reversible chemical bonding between fillers and matrices becomes imperative to integrate fillers into the healing chemistry effectively. hBN is known for its chemical stability, making it resistant to chemical alterations [176]. However, with advancing chemistry, techniques have been developed to introduce desirable functional groups onto the surface of the nanosheets. In an attempt to show the effect of functionalization, Jing et al. synthesized a nanocomposite hydrogel from PVA and –OH functionalized hBN nanosheets based on reversible hydrogen bonding. The resultant composite showed a much higher mechanical strength and also showed a 15% improvement in thermal conductivity by the addition of just 0.12% of hBN nanosheets [177].

When designing nanocomposites with enhanced thermal properties, it is imperative to understand the conductivity of fillers and matrices in different directions. For example, hBN exhibits anisotropic thermal conductivity, with significantly higher values along its basal plane compared to the perpendicular direction [178]. The in-plane thermal conductivity of 2D hBN can reach 1700–2000 W/m K, whereas the out-of-plane thermal conductivity is notably lower, typically in the range of 20–50 W/m K [179]. To fully harness the elevated in-plane thermal conductivity of hBN, Shang et al. strategically aligned the nanosheets with the direction of heat flow. This arrangement enhances the likelihood of energy propagation in the preferred direction [180]. The methodology combined an SH PDMS elastomer matrix reliant on dynamic imine bonds with BNNSs. Remarkably, the resulting SH PDMS composite incorporating 30 wt.% BNNSs showcased an impressive SH efficiency exceeding 85% and a remarkable 588% thermal conductivity enhancement relative to the pristine SH PDMS elastomer.

Although 2D hBN has shown promise in strengthening and SH of composites, Gao et al. have taken a step further by utilizing 1D porous hBN nanofibers [181]. These nanofibers offer flexibility that supports the movement of PVA polymer chains during the healing process. With the addition of just 1.25–2.75 wt.% hBN nanofibers, the hydrogels exhibited a remarkable 1080% increase in compression modulus and a 212% increase in compression strength. These nanofibers possess a high aspect ratio and an abundance of hydroxyl functional groups, making them active inorganic fillers that actively participate in the healing reaction, thereby enhancing the mechanical and SH properties of the composites. This work not only demonstrates the potential of hBN as a filler in composites but also highlights the importance of aspect ratio in improving properties. This concept can be translated into the area of 2D materials, where nanosheets characterized by a higher aspect ratio can demonstrate superior performance compared to nanosheets with a lower aspect ratio, particularly in terms of enhancing strength and healing capabilities.

Depending on the specific application and the ability to apply heat during use, alternative methods such as utilizing the reversible DA mechanism can be employed for enhanced and expedited healing, particularly when heat is available as a stimulus [182]. This approach offers quicker and more effective healing. In such instances, hBN emerges as an ideal filler material owing to its innate high thermal conductivity, facilitating efficient dispersion of heat throughout the matrix, thereby further augmenting the healing process [183]. In a study by Cao et al., hBN was modified through the utilization of a silane coupling agent to augment the compatibility between the hBN nanosheets and the epoxy matrix that served as the host material [184]. The incorporation of these modified hBN nanosheets resulted in a remarkable 2× enhancement in thermal conductivity when compared with the unmodified epoxy resin. The exceptional SH characteristics observed in the newly engineered composites were ascribed to a nearly ideal SH mechanism driven by the reversible DA chemistry, complemented by the assistance of shape memory effects. The research team extended the application of the hBN filler, which had been previously modified with maleimide functional groups [185], by incorporating it into an epoxy system featuring furan and epoxide functional groups, resulting in the creation of anticorrosive coatings. These coatings exhibit outstanding corrosion resistance, rapid healing capabilities achieved by heating at 120°C for 2 h, and effortless removal when needed. The optimum quantity of nanofiller in the polymer matrix needs to be identified; otherwise, it can have a detrimental effect on the performance as shown in Figure 9.

### 4.3 | MXenes

MXenes are a unique family of 2D materials very well known for their high strength, electrical conductivity, and tunable surface properties [186]. Structurally, MXenes are 2D structures with octahedral interlayers of carbon and/or nitrogen atoms nestled between transition metal layers [187]. More often, MXenes are synthesized by top-down approach by selectively etching out a specific atomic layer from a layered MAX phase [188]. Due to the wet chemistry process employed and the high surface energy of MXenes, they retain a plethora of surface functional groups that facilitate chemical bonding with the polymer matrix [189]. These 2D materials, endowed with excellent electrical and thermal properties, play a pivotal role in imparting or improving the SH properties of the PNCs. They not only provide active sites for reversible bond formation with the polymer matrix but also open additional pathways for healing by diverse stimuli such as electricity, heat, or light. In a very interesting work by Ge et al., it was also found that by configuring the molecular interactions between the polymer host matrix and MXene nanosheets, these 2D materials can act as crosslinkers and enable rapid gelation in a variety of polymers and monomers [190]. In a previous work, it was also found that the type of surface terminations can affect its healing and mechanical performance in an H-bond rich polymer matrix [191]. In recent years, research on self-healable MXene–polymer composites has progressed a lot, and it is no surprise that a wide range of applications have been identified by exploiting different properties of MXenes, as explained in the following section.

#### 4.3.1 | EMI Shielding

MXenes are endowed with superb electrical conductivity and a layered structure, which makes them a favorable choice for EMI shielding applications [192]. With 2D MXenes existing in a variety of combinations and surface properties, they can possess electrical conductivities ranging from 5 to over 20 000 S/cm depending on the type [193, 194]. Along with the intrinsic properties, the layered structure of 2D MXenes is responsible for effective energy attenuation of EM waves, making their performance much better than synthetic materials and almost comparable to traditional metal foils. There has been an increasing demand for multifunctional SH materials with effective EMI shielding in recent years [195]. When MXene is dispersed into a polymer matrix, it can be distributed uniformly and formed into a continuous conductive network that confers the necessary mechanical stability and lightweighting opportunity while retaining the shielding effectiveness [196, 197].

In one of the earliest studies exploring SH MXene-PNCs with EMI shielding capabilities, Nicolosi et al. developed a highly recyclable and stretchable $Ti_3C_2T_x$-PAA hydrogel using a biomineralization-inspired assembly method [198]. This composite, at a slim 130 μm thickness, demonstrated impressive EMI shielding efficiency (SE) of 45.3 dB over a wide spectrum range from 0.2 to 2.0 THz. In another study, when $Ti_3C_2T_x$ MXene was applied via dip coating within a PU foam matrix, the EMI SE skyrocketed to a remarkable 90 db [199]. This exceptional performance was attributed to the inherent porous structure of PU foam and the formation of shielding capsules, allowing such high efficacy with a mere 0.82 vol% MXene loading. To assess its SH capabilities, the healing efficiency, determined by the ratio of EMI SE after and before healing, was found to exceed 80%, providing a valuable measure of its overall performance. In another work, Fu et al. drew inspiration from dragonfly wing microstructures to create an MXene-supramolecular polymer composite using 2-ureido-4[1*H*]-pyrimidinone that strengthens brittle, healable polymers (25× increase in flexural strength), making them tougher (54.3× increase in fracture toughness) and more stable [200]. The material also offers improved thermal stability, remote NIR-triggered healing, and effective EMI shielding.

Beyond enhancing EMI shielding, MXenes also contribute positively to the mechanical properties of polymers. For instance, Jiao et al. substantially increased the tensile strength and toughness of WPU/natural rubber (NR) composites by adding just 5.48 vol% of $Ti_3C_2T_x$ [201]. These composite films, at a thickness of 339 μm, achieved an EMI SE surpassing 76.1 dB and exhibited a remarkable 90% healing efficiency following structural damage. In other cases, MXenes are integrated with other materials into the polymer matrix to bestow multifunctional properties. Wang et al. crafted a transparent conducting electrode by depositing AgNWs and $Ti_3C_2T_x$ MXene onto a 7,12-dimethylbenz(a)anthracene (DMBA)-modified oxime-carbamate PU film [202]. This composite boasted an 82.8% transmittance, a low sheet resistance of 18 Ω/sq, an EMI SE of 27.1 dB, and an impressive healing efficiency of 95%. Interestingly, $Ti_3C_2T_x$ MXene was combined with liquid metal within a WPU matrix to facilitate healing via joule heating and enhance EMI SE performance [203]. This resulted in an ultra-thin SH film with effective EMI shielding properties and exceptional healing efficiency. Likewise, an aligned 3D network of MXene and rGO was created through unidirectional freezing, with PU featuring DA bonding as the matrix [204]. This combination yielded an EMI SE of 39.1 dB and remarkable healing efficiency of 91.4%

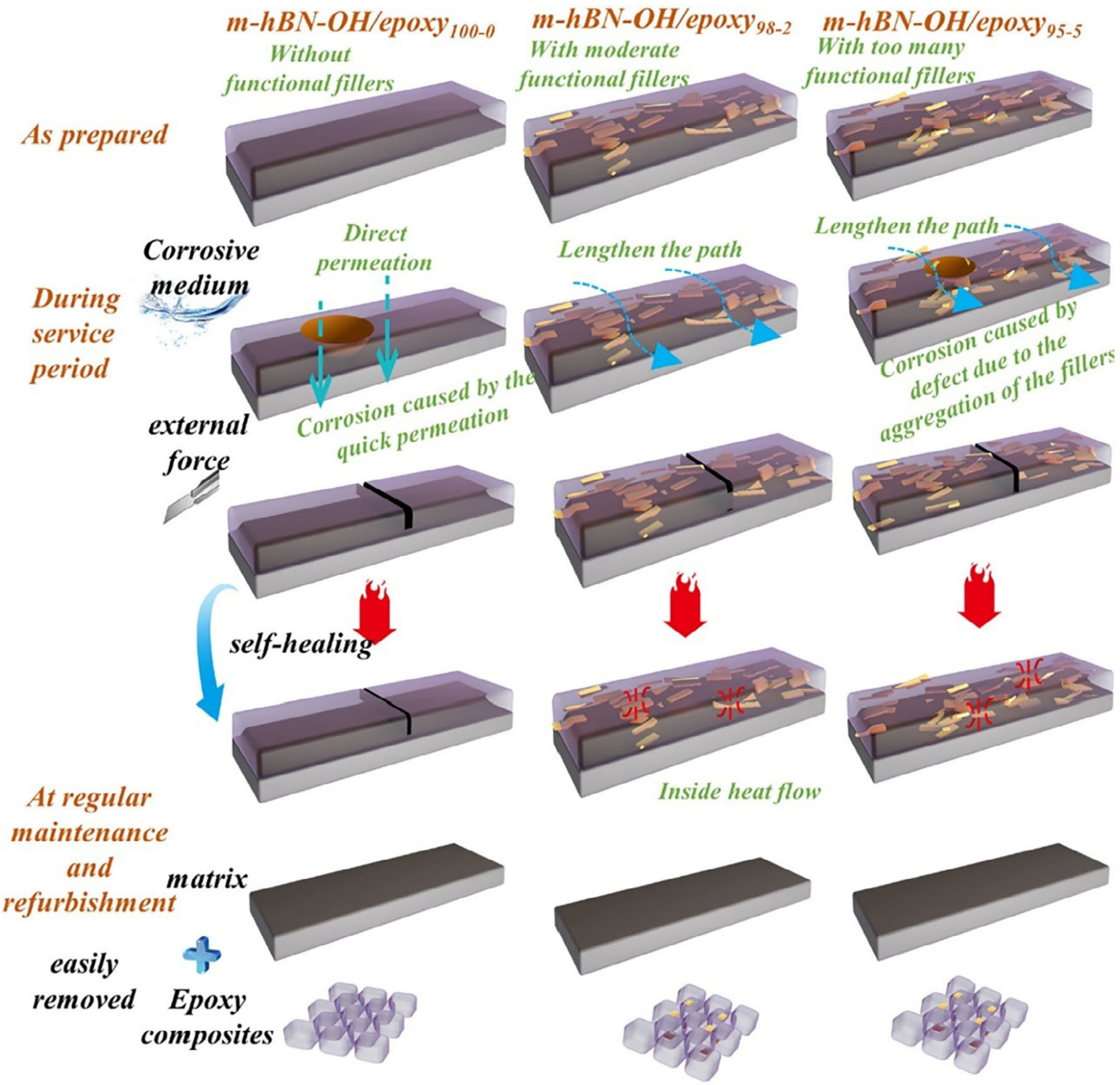

**FIGURE 9** | Illustration of the working mechanisms for hBN/epoxy composites at different loadings [185]. *Source:* Reproduced/Adapted from *Progress in Organic Coatings* [185] with permission from Elsevier.

with just 0.46 vol% of MXene and 0.65 vol% of rGO. As MXene composites continue to evolve as EMI shielding materials with such high performance at low loading, they offer the potential to revolutionize a broad spectrum of industries, from telecommunications to aerospace, where EMI shielding and lightweighting are of paramount importance [205].

### 4.3.2 | Energy Harvesting and Storage

MXene-based SH polymer composites promise a significant enhancement in the performance, durability, and longevity of energy devices to address the global transition towards cleaner and more sustainable energy sources. Utilizing their innate high electrical conductivity, MXenes have been applied in the fabrication of supercapacitors through the incorporation of electrically conductive polymer hydrogels (ECH) [206]. This integration yields electrodes possessing conductivity akin to that of metals. Additionally, the extensive surface area of MXene nanosheets facilitates easy electrolyte penetration, with MXene displaying minimal interference in charge transfer processes. These electrodes not only offer SH at room temperature, flexibility, and high tensile stress of 0.083 MPa, but they can also achieve a maximum capacitance of 291.8 mF/g at 0.5 mA/g and maintain 64.24% (healing efficiency) capacitance retention after 2000 charge–discharge cycles.

In response to the energy storage needs of nano/micro devices, Yue et al. have crafted high-performance microsupercapacitors (MSCs) using an MXene-rGO 3D aerogel enclosed in an SH PU casing [207]. This hybrid design maintains 81.7% of its initial capacitance even after undergoing five healing cycles. In a related development, Li et al. engineered PU-encased MXene-based electrodes combined with a PVA/$H_2SO_4$ electrolyte to create healable MSCs [208]. The MXene electrodes were fabricated through printing and laser engraving techniques. The PU component contributes by providing essential mechanical stability and SH properties to the overall structure. Although the MXene nanosheets themselves do not actively participate in the healing process, the SH properties of the PVA/$H_2SO_4$ electrolyte and the PU outer layer ensure that these manufactured MSCs retain 90% of their capacitance after 2000 charge–discharge cycles, even following five instances of cutting and healing at the same site. Utilizing a similar MXene-PU aerogel framework, Hu et al. achieved outstanding thermal energy storage capabilities by encapsulating phase-changing materials (PCMs) [209]. The addition of MXenes enhances photo absorption capabilities, PU supports healing properties, and the PCMs are responsible for light-triggered shape memory functions. Further, in the domain of energy harvesting, triboelectric nanogenerators (TENGs) excel at the effective utilization of mechanical energy. They can employ this energy for self-powering the material, which also serves as a sensor, or for general energy harvesting [210]. TENGs operate through the interaction of triboelectric materials, achieved via contact electrification and electrostatic induction, resulting in a synergistic and efficient effect. He et al. developed a flexible TENG composite designed in layers using MXene, which exhibits impressive SH capabilities [211]. This SH can occur at a temperature around 37°C within a mere 4 h or, for faster healing, using a hair dryer in just 5 s. This composite, boasting a healing efficiency of 98%, not only showcases its prowess in energy harvesting—evident through its capacity to generate a peak-to-peak voltage of 170 V and a current of 160 μA when periodically compressed by a human hand—but also excels in sensing, accurately tracing the position touched by a finger.

### 4.3.3 | Protective Coatings

As with other 2D materials, MXenes are also known for their outstanding properties when used as passive physical barriers in polymer composites. They create a tortuous path in the host matrix that is highly impermeable to ions and provides an effective barrier against water and oxygen [212]. This is achieved by forming a complex labyrinth-like structure within the composite matrix, like other 2D materials, which significantly lengthens the path that water and oxygen must traverse to reach the protected substrate [213]. Additionally, introducing SH properties to these coatings ensures that even when the surface is mechanically damaged, such as by scratching, the environmental damage can be minimized and halted as the coating repairs itself. The corrosion resistance of MXene-based polymer coatings depends not only on the intrinsic properties of the MXenes but also on factors such as the nanomaterial content and coating thickness. An optimal balance is required to maximize protection. Increasing MXene content can enhance the barrier properties by introducing more tortuous paths, which slows ion diffusion. However, excessive content may lead to particle aggregation, reducing coating uniformity and weakening corrosion resistance. Similarly, coating thickness plays a critical role: Although thicker coatings offer better physical protection, they may suffer from reduced flexibility or adhesion, impacting their durability in practical applications. In one of the works, Li et al. synthesized an anticorrosion coating using methionine functionalized $Ti_3C_2T_x$ nanosheets in a PU matrix that can be healed at room temperature after being damaged as shown in Figure 10 [214].

This coating uses a combination of active and passive protection to prevent corrosion of the surface. When the coating sustains damage, methionine corrosion inhibition attaches itself to the metal surface's anode site by adsorption and slows down the anodic reaction. The presence of dynamic disulfide bonds and H-bonds promotes scratch closure. When tested for Mg-alloys, the intrinsic SH mechanism and active/passive protection strategy of the coating translate into exceptional, long-lasting corrosion resistance. When the healing efficiency of the composite coating in terms of anticorrosive properties was measured, it was found to be 140%, mainly due to the adsorption of methionine on the exposed surface.

MXenes are often used along with other nanomaterials to impart multifunctionality to SH coatings. For instance, an epoxy-based composite comprising MXene-$SiO_2$ was tested for its anticorrosion properties and exhibited a healing efficiency of 91% [215]. In this case, $SiO_2$ serves as a nanocontainer that holds tannic acid, a corrosion inhibitor that is released upon mechanical damage. In another example, a coating was developed with antifouling, anticorrosive, and SH properties by incorporating $Ti_3C_2T_x$ MXene along with the ZIF-8 metal–organic framework (MOF) into a PU matrix [216]. When damaged, this coating releases Zn ions and mercaptobenzothiazole, leading to a reduction of 99% in surface bacteria and 93% in microalgae. Similarly, an epoxy coating containing $Ce^{3+}$ doped MXene nanosheets was also designed, with $Ce^{3+}$ serving as a corrosion inhibitor that is released when the coating is damaged [217]. Depending on the healing chemistry of the polymer matrix, MXene-based coatings can also be tuned to enable spot healing through the use of NIR [218] and solar light [219].

### 4.3.4 | Biomedical Applications

Adhesive hydrogels, valued for their functional attributes, have generated significant interest owing to their wide-ranging applications, especially in the realms of smart electronic skins and flexible biomedical devices. However, they still require improvement in mechanical properties, which can be achieved by incorporating various 2D materials, like MXene. In one notable study, a highly adhesive and ionically conductive hydrogel was synthesized through $\gamma$-radiation polymerization of 2-(dimethylamino)ethyl methacrylate (DMAEMA) along with a low concentration of MXene nanosheets [220]. The MXene in this research serves a dual purpose, enhancing both the hydrogel's conductivity and strength, while also playing a critical role in achieving a uniform polymer structure. Similar expedited gelation effects were observed when $Ti_3C_2T_x$ nanosheets and liquid metal nanodroplets were used to create soft electronic materials for sensing, force mapping, and electroluminescence applications using bacterial nanocellulose and PAA hydrogels [221]. This was accompanied by an SH efficiency of nearly 97.6% (mechanical) and 100% (electrical) directly facilitated by a dynamic hydrogen bonding network. Highlighting the potential of biopolymers, an MXene-based hydrogel was developed using chitosan and honey, showcasing biocompatibility with various stem cells and natural antibacterial and anti-inflammatory properties [222]. The implications of this development are particularly exciting for applications in regenerative medicine, offering substantial potential for tissue repair in the context of degenerative diseases. Furthermore, in a notable study underscoring the promise of

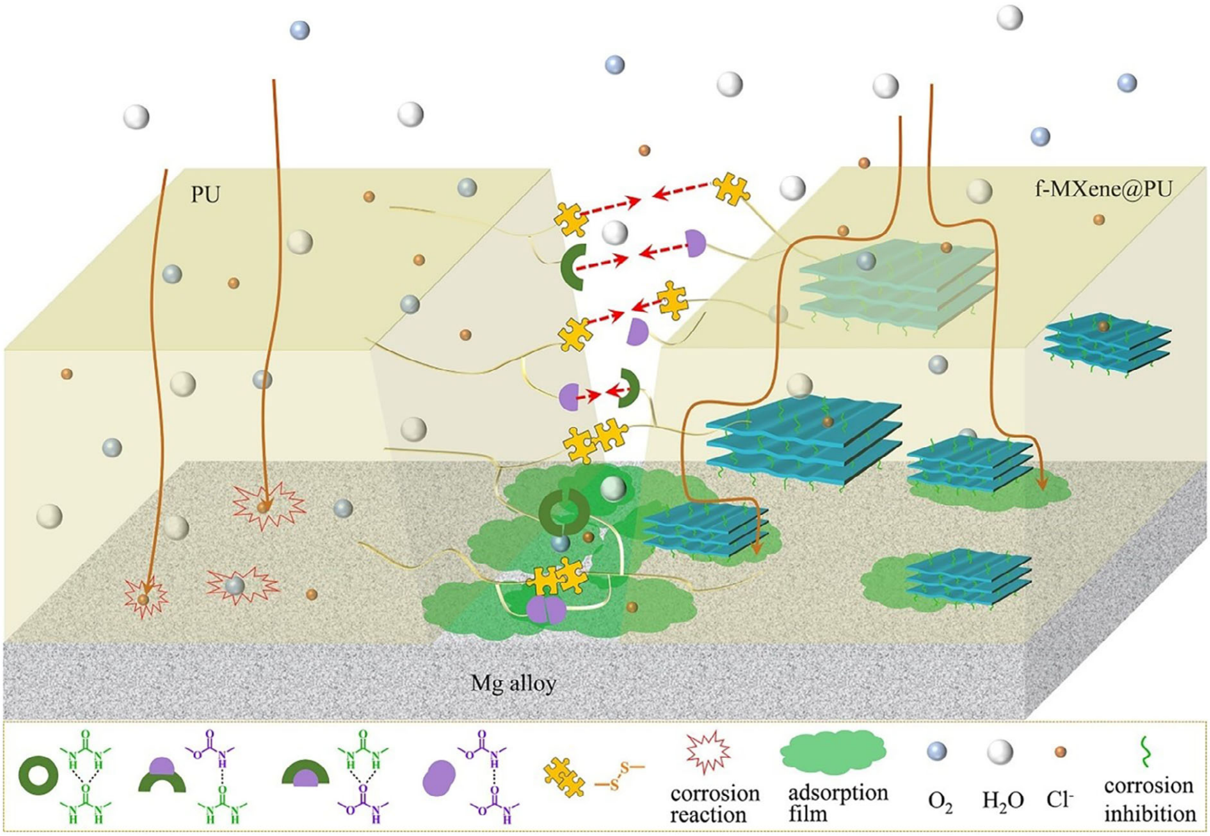

**FIGURE 10** | Anticorrosion/SH mechanism of MXene@PU coating [214]. PU, polyurethane. *Source:* Reproduced/Adapted from *Chemical Engineering Journal* [214] with permission from Elsevier.

biopolymer-MXene composites, Zhu et al. harnessed dynamic imine bonding to produce oxidized alginate and gelatin hydrogels incorporating MXene [223]. This development has potential applications in wound healing and bioelectronics. However, this work does not quantify the material's SH properties, making it challenging to determine the extent of property recovery after healing.

Multifunctionality is one of the main features when the composite is intended to mimic human organs. In a recent work by Xu et al., stretchable thermoconductive skins were developed using poly(urea–urethane) (PUU) for thermal dissipation in robotic joints [224]. This was enabled by high in-plane thermal conductivity of MXenes and excellent reinforcing effect. The resulting composite possesses record-high fracture energy at 501.6 kJ/m$^2$ with 97% healing efficiency. Similar good progress has been done in SH TENG materials, which possess the capability to monitor and harness energy from human motion and are considered strong contenders for the development of an ideal e-skin. To explore this further, Cai et al. engineered a composite comprising $Ti_3C_2T_x$ MXene and PDMS, which was reinforced through a dual crosslinking strategy by fine-tuning the ratio of H-bonds to imine bonds [225]. This unique material offers adaptability to conform to the irregular surface of human skin, maintaining its TENG device performance even after enduring multiple cycles of damage and repair. Furthermore, it boasts a highly stretchable structure and exhibits the remarkable ability to self-heal at room temperature, with a healing efficiency of 78%.

### 4.3.5 | Wearable Sensors

The suitability of MXenes as conductive 2D nanofillers for polymer-based sensors is well-established, as evidenced by the substantial body of research in this area [226–230]. Similar to other 2D materials, MXene-based sensors operate on the principle of changes in the electrical conductivity of percolating 2D particles embedded within an insulating matrix [231, 232]. MXenes contribute highly to such nanocomposite sensors by providing mechanical strength, durability, linear response over a wide strain range, and the ability to recover sensing ability after mechanical damage [232]. For instance, Liao et al. engineered an innovative antifreeze and SH MXene-based organohydrogel exhibiting exceptional strain sensitivity even at ultra-low temperatures [233]. This was accomplished through a two-step process: firstly, crosslinking PVA with borate ions using a freeze-thaw method, and second, in situ polymerization at 60°C to form an interpenetrating PAM network. To enhance the organohydrogel's antifreeze properties, partial solvent displacement was employed by immersing the hydrogel in ethylene glycol, effectively replacing a portion of the water molecules. The resulting organohydrogel demonstrated a remarkable SH efficiency of 85%, attributed to the abundance of hydrogen bonds that facilitated the restoration of mechanical properties. In the development of MXene-based nanocomposite sensors, a critical challenge lies in optimizing MXene loading. Although higher MXene content increases the likelihood of forming continuous pathways for electron mobility, it simultaneously impedes the segmental motion of polymer chains, thereby compromising healing efficiency. Furthermore,

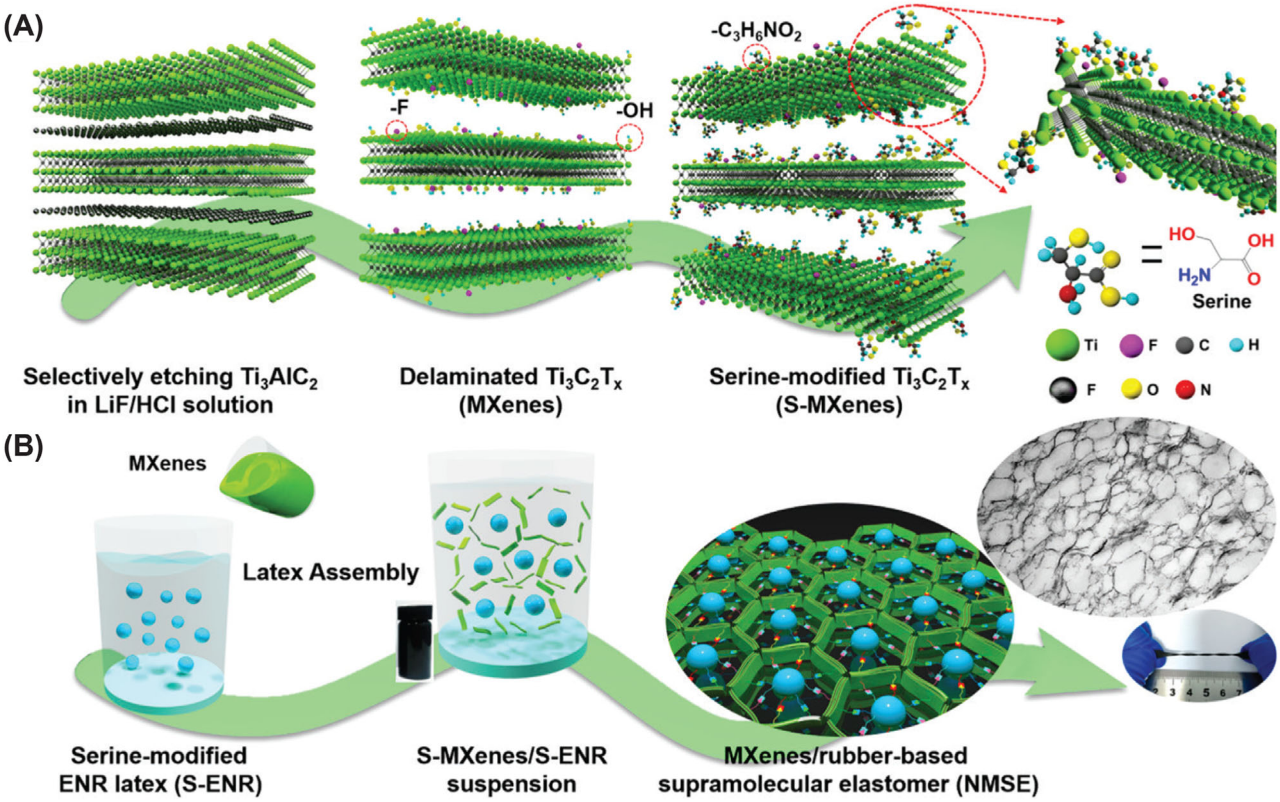

**FIGURE 11** | Main preparation process for NMSE. (A) Surface modification of MXene nanosheets by serine via the esterification reaction. (B) Construction of a nanostructured MXene network in NMSE via the latex assembly method [234]. *Source:* Reproduced/Adapted from *ACS Nano* [234] with permission from American Chemical Society.

excessive MXene loading can adversely affect the flexibility and stretchability of the nanocomposite. Guo et al. found an answer to this and harnessed the self-assembly properties of polypeptides to fabricate a highly sensitive and self-healable nanocomposite capable of detecting subtle strains induced by facial expressions, speech signals, and human pulses [234]. The nanocomposite's properties stem from the synergistic combination of serine-functionalized MXene nanosheets and epoxidized rubber as shown in Figure 11. The serine functionalization promotes the self-assembly of MXene nanosheets, imparting robust structural integrity and efficient stress distribution. Meanwhile, the incorporation of epoxidized rubber imbues the nanocomposite with exceptional SH capabilities, achieving a healing efficiency of 99.34% and a remarkable strength of 4.55 MPa. Notably, the nanocomposite exhibits a remarkably rapid response time of 50 ms and a high gauge factor of 107.43, underscoring its potential for real-time strain sensing applications. This was attributed to the synergistic effects of hydrogen bonding and self-assembly, enabling effective network formation at a remarkably low MXene loading of 6 wt.%. In contrast, randomly distributed MXene required a significantly higher loading of 40 wt.% to achieve a percolation network, highlighting the crucial role of self-assembly in optimizing material utilization and performance.

Another prevalent issue encountered in MXene-based sensors is the manifestation of permanent hysteresis following multiple healing cycles. With each successive healing cycle, resistivity tends to increase marginally due to imperfect surface connections or incomplete healing. In cases where spontaneous bonding mechanisms such as hydrogen bonding are involved, bond formation on the same side of the surface can also contribute to conductivity loss. This can be minimized by introducing highly dynamic functional groups by surface functionalization that can assist in bond formation with the polymer matrix and minimize the hysteresis [235]. A summary of MXene-based sensors can also be found in Table 1.

## 4.4 | Clays

Clay nanosheets have been investigated as promising additives for SH polymer composites [256]. This is mainly because of their ion-exchange capability, which simplifies functionalization, high aspect ratio, and high stability in H-bond rich medium [257, 258]. Clays also offer the possibility to adjust transport characteristics, aiding in the insertion of molecules or the creation of nanoporous structures within the clay [259]. Moreover, certain clays possess inherent SH properties, enabling their particles to restore mechanical, rheological, and conductive characteristics, which can be further enhanced through mild polymer intercalation [260, 261]. Among these, the montmorillonite (MMT) type stands out due to its affordability, excellent intercalation properties, chemical stability, and good electronic transfer characteristics [262]. MMT has found application not only in SH polymers but also in the improvement of healable concrete [263] and asphalt mixes [264]. In a study by Gao et al., acrylamide (AM) was polymerized in the presence of exfoliated MMT nanosheets using in situ polymerization [265]. This resulted in composites

**TABLE 1** | Comparison of different MXene-based polymeric sensors.

| Polymer matrix | MXene functionalization | Bonding mechanism | Healing stimuli | MXene loading | Mechanical strength | SH efficiency | Response time (ms) | Guage factor | References |
|---|---|---|---|---|---|---|---|---|---|
| PDMS | Thioctic acid | Hydrogen and metal–ligand bonds | RT | 12 | 2 | 91.7 | NA | NA | [236] |
| PU + aramid nanofibers | — | Hydrogen bonds | RT | 50 | 114 | NA | 10 | NA | [237] |
| Poly(*N*-isopropylacrylamide) | — | Hydrogen bonds | RT | 0.1 | 0.4 | 76.1 | NA | NA | [238] |
| PAM + Sodium alginate | Polydopamine | Imine and hydrogen bonds | RT |  | 0.17 | 91 | 263 | 2.2 | [239] |
| PVA + PAA | — | Hydrogen bonds | RT | 3 | 0.035 | 85 | NA | 44.85 | [233] |
| Sodium alginate + PAM | — | Hydrogen bonds | RT |  | 0.014 | 88.75 | 200 | NA | [240] |
| PDMS | D-Asparagine | Imine and hydrogen bonds | RT | 10 | 1.81 | 98.4 | NA | NA | [235] |
| PDMS | L-Citrulline | Disulfide and hydrogen bonds | RT | 10 | 3.65 | 91 | NA | 3.38 | [241] |
| Polysaccharides | Sulfanilic acid diazonium | Hydrogen and ionic bonds | RT | 0.3 | 0.06 | ≈90 | NA | 1.1 | [242] |
| Acrylate vitrimer | — | Hydrogen bonds | Microwave | NA | 0.13 | 90 | NA | 3.71 | [243] |
| Poly(MPTC-NaSS) | — | Hydrogen and ionic bonds | RT | 1 | 1.48 | 86 | NA | 6.32 | [244] |
| Agar | — |  | RT/Heating to 100°C | 0.3 | 0.129 | 25.9/100 | NA | 1.44 | [245] |
| PEA | — | Diels–Alder and hydrogen bonds | Heat | NA | 11.22 | 88 | 71 | 29.4 | [246] |
| Poly (ionic liquid) | — | Covalent and hydrogen bonds | RT | 2 | 0.21 | 77.93 | 100 | NA | [247] |
| PVA | — | Covalent and hydrogen bonds | RT | 50 | 0.0097 | 96.3 | NA | 2.3 | [248] |
| Poly(ethylene-vinyl acetate) | — |  | Heating | NA | 31 | 94 | 178.9 | 82.5 | [249] |
| PAA + bacterial cellulose | — | Hydrogen bonds | RT | NA | 0.17 | 100 | NA | 8.09 | [221] |
| Poly(AM-*co*-AA) + chitosan | — | Hydrogen bonds | RT | 0.3 | 0.003 | 45.3 | NA | 3.93 | [250] |
| PVA + PAA | CMC + sodium tetraborate | Hydrogen and borate-ester bonds | RT | 3.2 | 2.24 | 84 | 13 | 1.11 | [251] |
| PVA | — | Hydrogen and borate-ester bonds | RT |  | 0.75 | 91 | 150 | 3.26 | [252] |
| PAA | — | Hydrogen | RT | 0.12 | 0.18 | 100 | 20 | 10.79 | [253] |
| Epoxidized natural rubber | Serine | Hydrogen bonds | RT | 6 | 4.55 | 99.34 | 50 | 107.43 | [234] |
| Poly(AA-*co*-SMA) | — | Hydrogen bonds | RT | 32.4 | 0.525 | 98 | 79 | 2.2 | [254] |
| Poly (ionic liquid) | (3-Aminopropyl) triethoxysilane | Hydrogen and ionic bonds | Heat | 10 | 11.45 | 90 | NA | NA | [255] |

Abbreviations: AA, acrylic acid; AM, acrylamide; CMC, carboxymethyl cellulose; MPTC–NaSS, [3-(methacryloylamino)propyl]trimethylammonium–sodium *p*-styrenesulfonate; NA, not available; PAA, polyacrylic acid; PAM, polyacrylamide; PDMS, polydimethylsiloxane; PEA, poly(propylene glycol) bis(2-aminopropyl ether); PU, polyurethane; PVA, polyvinyl alcohol; SMA, stearyl methacrylate.

displaying fracture elongation of 11,800% and fracture toughness of 10.1 MJ/m$^3$. Under cyclic loading tests, these composites exhibited the recovery of residual strain under mild conditions, attributed to the reinforcement by MMT nanosheets. The primary mechanism identified was based on the reversible desorption and adsorption of polymer chains on the surface of the clay platelets. Using a similar in situ polymerization process, macromolecular structures from xylose-based hydrogels [266] and PU-acrylate [267] were also synthesized using inorganic and organic MMT, respectively. Modification of MMT using cationic inhibitors has also been investigated to improve its performance as an additive for SH composite coatings designed for corrosion protection. This is usually done by coercing chemical complexation of corrosive ions using rare earth metal ions. For example, Ce$^{3+}$ and tannic acid along with MMT have been used to improve the anticorrosive properties of self-healable WPU and improve the impedance modulus by three orders of magnitude as compared to virgin polymer coating [268]. Similarly, Ce$^{3+}$ and Zr$^{4+}$ ions were used to improve the charge transfer resistance and corrosion protection of organosilicon polymer matrix to protect aluminum alloys [269]. Several clays, characterized by layered structures such as zeolite [270], bentonite [271], laponite [272–274], mica [275–277], and even those with a 1D structure like sepiolite [278], have been explored as fillers for SH polymer composites. However, in-depth investigations into their specific sheet properties, influence on reinforcement mechanisms, structure–property relationships, and compatibility with other intrinsic healing chemistries have not yet been documented.

In a study by Post et al., MMT has also been explored as an additive for extrinsic healing of compartmented glass fiber-based composites where healing is fostered by epoxy- and thiol-based chemistries [279]. This research is a crucial step in enhancing the recovery of matrix properties in fiber-reinforced composites, where restoring properties after fiber damage is highly challenging. The analysis of mechanical properties indicated that compartmented fiber technology ought to prioritize healing matrix damage exclusively, given that restoring mechanical integrity after glass fiber fracture is insurmountable.

Layered double hydroxides (LDH) are a class of material from the anionic clay family exhibiting 2D brucite-like cationic structures interspersed with charge-compensating anions and solvation molecules in between the layers. The edge-connected octahedral structure of LDH contains metal cations as the center, and the vertices of the octahedral structure contain hydroxide ions, connecting to form 2D sheets with infinite lateral dimensions. LDHs have a general formula of $[M_{1-x}{}^{2+}M_x{}^{3+}\ (OH)_2]^{x+}[A_{x/n}]^{n-}\cdot mH_2O$ where $M^{2+}/M^{3+}$ is a divalent/trivalent metal cation and $A_{x/n}$ represents the charge compensation brought in by anionic moieties within the interlayer gallery [280]. The addition of LDH to polymer matrix has been shown to bring in great improvements in the thermal properties, flame retardancy, and overall physical properties of the composites [281]. Increasing the adhesion between LDH and the polymer matrix can provide enhanced mechanical strength to the composite with the possibility of instilling SH ability. It is essential that the bonding induced on the surface is not irreversible and that it is possible to reconnect the bonds with minimum external energy [256]. LDHs have been previously functionalized by different strategies such as layer composition tuning [282], surface modification [283], hybrid assembly [284], intercalation [285], size and morphology regulation [286], and defect introduction [287]. Earlier attempts were made to synthesize SH microcapsules for anticorrosion applications using LDHs, where LDHs were part of the SH composite but did not actively participate in the healing chemistry, and they were rather used to serve secondary purposes such as chloride ion leaching [288, 289]. Similarly, LDHs have also been used in conjunction with other 2D materials like graphene [290], rGO [291], and MXene [292] in separate cases to synthesize multifunctional SH polymer coatings. Sometimes, 3D nanostructures like MOFs can be used with LDHs to enhance the resistance to high and low-pH chemical corrosion. Using this, self-repairing anticorrosion polymeric coatings have been developed by incorporating LDHs decorated with MOFs, further elevating their chloride adsorption, multifunctionality, and resistance to corrosion [293, 294]. Many of these LDHs can be directly sourced from nature and exfoliated with cost-effective methods. Their notable active surface, characterized by hydroxyl terminations capable of both accepting and donating hydrogen bonds, positions them favorably for use as 2D additives in SH polymeric composites. Acknowledging that LDHs can lower the crystallization temperature of polymers [295], increasing the likelihood of crystallization during the healing process and potentially diminishing post-healing strength due to interfacial crystallization, it is crucial to note that despite providing additional bonding sites, the hydroxyl termination elevates the surface energy of the nanosheets. Despite the abundance of bonding sites, there has been a lack of exploration into surface functionalization of the LDH nanosheets to introduce additional bonding mechanisms, such as DA reactions or Schiff-base reactions. This unexplored avenue could potentially leverage flame retardancy, thermal, and mechanical properties at healing temperatures within the composite. Moreover, the increase in surface energy brought in by hydroxyl terminations could be tuned to make it selectively compatible with the host polymer matrix.

### 4.5 | Other 2D Materials

Besides graphene, TMD, hBN, MXene, and clays, numerous additional 2D materials have lately been explored for SH polymer composites owing to their varied structures and characteristics. Graphitic carbon nitride (g-$C_3N_4$) adopts a graphite-like structure with sp$^2$-hybridized carbon and nitrogen atoms, where some carbon atoms are replaced by nitrogen atoms [296]. It can exist in *s*-triazine-constructed or tri-*s*-triazine-constructed configurations and can be synthesized by both bottom-up and top-down methods [297, 298]. Using the excellent strength, flexibility, and load-bearing capacity of g-$C_3N_4$, Xu et al. designed a self-healable nanocomposite by a polycondensation reaction between poly(propylene glycol), tolylene 2,4-diisocyanate terminated, and adipic dihydrazide (AD) [299]. It was also crosslinked with g-$C_3N_4$ NSs via nucleophilic addition and rearrangement reactions. g-$C_3N_4$ acts as a reinforcing nanofiller, a barrier to slow penetration of aggressive corrosive species, and also improves moisture sensitivity of hydrogen-bonded macromolecular structures. This PUU–g-$C_3N_4$ smart coating offers excellent corrosion protection and healing properties even at very high humidity. g-$C_3N_4$ nanosheets have also been tested for use in healable PAM hydrogels for UV shielding applications [300].

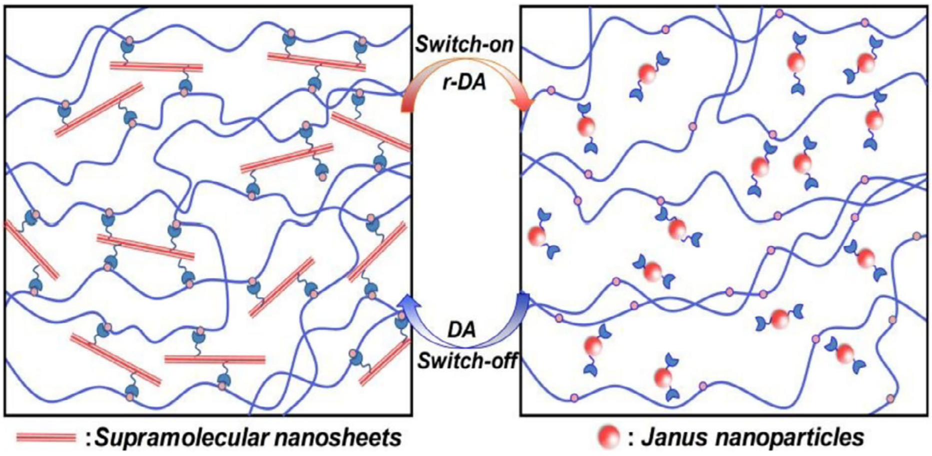

**FIGURE 12** | Schematic for the switchable polymer hybrid network composed of the PU and supramolecular nanosheets [309]. *Source:* Reproduced/Adapted from *Journal of Materials Chemistry A* [309] with permission from Royal Society of Chemistry.

Depending on the properties desired in the composites, other inorganic 2D materials, such as aluminum hydroxide [301], black phosphorus (BP) [302], $BiMoO_6$ [303], metal nanosheets [304, 305], and even silica-based Janus nanosheets [306, 307], have been tested for SH polymer composites. There are also some other 2D materials that have been synthesized, and their fundamental characteristics have been comprehended such as graphene, borophene, germanene, silicene, $Nb_3X_8$, and $FeB_2$ [308]. However, their potential application in SH composites and their capacity to enhance multifunctionality within composite systems remain largely unexplored. Investigating these materials further, as well as delving into other members of this expansive group of 2D materials based on how they interact with polymers and what added value they could bring, presents a promising opportunity for fundamental scientific research.

The inclusion of 2D fillers often becomes a requisite measure aimed at offsetting the loss in mechanical properties of polymers designed with healable characteristics. This decline arises from the inherent demand placed upon polymer chains within SH polymers, necessitating swift dynamics to enable seamless network reconfiguration following instances of fracture. Yet, this trade-off results in the constriction of chain mobility due to the physical obstruction imposed by these significantly expansive 2D fillers. To address this, Wei et al. synthesized hep-isobutyl polyhedral oligomeric silsesquioxane (POSS)-linked supramolecular nanosheets of 1,3,5-benzenetriamides core with maleimide and alkyl terminations as shown in Figure 12 [309]. During the DA-crosslinking process, the Janus nanoparticles self-assemble into 2D amorphous supramolecular nanosheets with an average thickness of 5 nm due to decreased solubility. At low temperatures, these nanosheets act as sturdy nanofillers, establishing robust interfacial DA bonds that enhance the elastomers' mechanical strength ("switch-off" state). Conversely, at higher temperatures, the dissociation of these bonds ("switch-on" state) drastically reduces crosslinking density. Consequently, the nanofillers disperse into a molecular state, boosting polymer mobility and facilitating effective macroscopic damage repair. On the basis of DA mechanism, this technique highlights a very important concept in healable nanocomposites where nanosheets can breakdown into a state of molecular dispersion, allowing for heightened polymer molecule mobility without hindering the chain mobility, thereby empowering the elastomers to effectively mend macroscopic damages. Taking advantage of such switchable interface connections provides a path ahead for novel healing chemistries working in synergy with the transformable structure of the fillers without compromising inherent polymer properties.

### 4.6 | SH Polymers Based On 2D Nanofiller Hybrids

Hybridization of 2D nanosheets with 1D [202], 2D [145], or 3D [310] nanofillers enables customization of each filler's characteristics, enhancing either the multifunctionality, healing characteristics, or reinforcement of polymeric nanocomposites. When 2D nanosheets interact with other fillers, they assume distinct roles: (1) They help stabilize other similarly sized 2D nanosheets by electrostatic interactions, (2) they can provide a scaffold for growth of 1D materials that can help in alignment of such 1D materials and improve axis-dependent intrinsic material properties such as conductivity and strength, and (3) 2D materials can be decorated with 3D nanoparticles, preventing agglomeration of 2D nanosheets while leveraging the advantageous multifunctional properties of 3D nanofillers, thereby improving dispersion within the composite structure. A variety of unique combinations have been investigated within hybrid structures, particularly 2D–2D hybrids such as MXene–LDH [292], TMD–perovskite [145], LDH–hBN [311], silica–GO [312], and MXene–graphene [207]. During the synthesis of SH polymers incorporating hybrid nanofillers, meticulous consideration should be exercised in the selection and customization of constituents based on their surface charge, functional groups, lateral size, and intended function within the composite, emphasizing a deliberate approach over arbitrary selection from a pool of nanomaterials. This approach facilitates the achievement of a true "synergistic effect" resulting from the incorporation of multiple nanomaterials, surpassing improvements solely governed by basic mixing principles.

## 5 | Characterization, Monitoring, and Healing Assessment

Material characterization of SH polymers with 2D materials gives a deeper insight into the specific roles played by 2D materials in property enhancement, identifies optimal material compositions, examines potential failure factors, and assesses the applicability of these findings within intended practical uses. This offers invaluable insights into the intricate interplay between SH mechanisms and the incorporation of 2D materials, shedding light on both the strengths and limitations of a particular composite material. The choice of characterization methods relies not solely on the properties of 2D materials but also on the specific traits exhibited by the base matrix. For instance, it is advisable to employ tensile tests and double cantilever tests when exploring the healing attributes within brittle polymers, whereas the assessment of fracture behavior stands as an optimal approach to grasp the healing properties in ductile polymers [313].

In the assessment of SH polymers, numerous studies have predominantly relied on direct visual observations using optical microscopy, often for qualitative evaluations of the healing process [66, 95, 314]. Techniques like optical profilometry [315] and confocal microscopy [316] have also been used to analyze scratch volumes and crack dimensions, albeit operating at length scales surpassing those necessary for studying micromechanical aspects involving 2D materials. To address this gap, scanning electron microscope (SEM) emerges as a valuable tool, offering higher resolution to explore the role of 2D materials at the healing interface. Furthermore, SEM facilitates the assessment of distribution, agglomeration, and reinforcement of these materials within the polymer matrix [98]. For more intricate analysis demanding finer resolution, transmission electron microscope (TEM) proves highly advantageous, particularly in systems incorporating multiple 2D materials or hybrid configurations involving diverse nanomaterials [317, 318]. The application of TEM becomes critical in studying composites wherein structural alterations in 2D materials occur during manufacturing processes, such as further exfoliation during mixing or nanosheet fracturing leading to variations in the structural properties. Atomic force microscope (AFM) also serves as a viable method not only to discern the uniform distribution of nanosheets but also to create nano-level scratches, enabling qualitative observations of healing in specific regions [319]. Furthermore, the integration of AFM with heating stages and additional accessories allows for in situ measurements, particularly in healable composites necessitating stimulation for healing. These techniques enable the observation of the mending process at the surface or close to the crack interface due to the opacity of the majority of polymers to both light and electrons. X-ray computed tomography is emerging these days as a valuable tool to observe the damage and healing processes in three dimensions, aiding in the comprehension of crack propagation mechanisms, nanosheet alignment, and subsurface restoration [227]. Despite the prevalent use of optical microscopy for qualitative analysis, a potential limitation arises when employing this method to study narrow scratches, especially in SH polymer composites requiring higher temperature healing. This may result in artificially narrowed or completely disappeared cracks due to thermal expansion, potentially misconstruing the observation as successful healing without actual material property recovery [320]. This could be aggravated in SH composites with thermally conductive nanofillers like graphene or hBN. Hence, emphasizing the need to complement visual observations with assessments of material properties or chemical changes occurring during the healing process becomes imperative for the accurate understanding of the healing extent in SH polymers.

Over the years, mechanical properties have served as crucial indicators to assess the healing capabilities of polymeric composites. After a nanocomposite is physically damaged, the end-user's primary concern lies in understanding the impact of these healing processes on bulk-scale mechanical properties regardless of the molecular processes or intricate details of the healing process. Consequently, evaluating these properties before and after healing using various tests, such as tensile, compression, bending, or impact, has become a prevalent approach to determining healing efficiency, represented by the ratio between pre- and post-healing strength parameters [320]. Although strength is commonly employed for comparisons, toughness offers a more comprehensive evaluation by encompassing the ductility parameter, providing a more accurate assessment [321]. As it can also be visualized, the healing performance varies across different tests. For instance, a compression test can generate the necessary mechanical force conducive to enhancing physical chain entanglement and fostering additional bond formation on the crack surface, showcasing higher efficiency in healing. Most studies in this domain employ custom-made samples with non-standard dimensions, posing challenges for direct comparisons. Researchers have explored numerous specimen geometries for SH polymers, such as double cleavage drilled compression specimens [322], double cantilever beams [323], tapered double cantilever beams [321, 324], compact tension specimens [325], single-notched beam specimens [326], fully constrained tensile specimens [327], anti-buckling compression after impact specimens [328], end-notched flexural specimens [329], and single-leg bending specimens [329]. Understanding these diverse specimen geometries is paramount, especially in material testing for SH composites incorporating 2D materials, given that many properties of 2D materials are axis dependent.

The healing process in SH PNCs predominantly occurs at the molecular level, with its effects manifesting at the bulk scale. Among the commonly preferred spectroscopy techniques to observe healing, Fourier-transform infrared spectroscopy (FT-IR) stands out, enabling the comprehension of healing through the formation of new bonds or the reformation of broken bonds after healing. This is discerned by assessing the peak intensity of relevant bonds, offering valuable insights into molecular-scale processes and avenues for enhancing bulk properties. For instance, Zeng et al. focused their FT-IR investigation on the 1700 and 1730 cm$^{-1}$ bands, corresponding to the carbonyl stretching vibrations in the maleimide crosslinker and the poly(2,5-furandimethylene succinate) chain, respectively. The study relied on the increased intensity of these peaks relative to the unreacted maleimide bond at 700 cm$^{-1}$ as an indicator of the reaction's progress [330]. Raman spectroscopy can also be used to characterize the healing dynamics [331], distribution of 2D materials in the matrix [43], and even understand strain transfer in composite materials by testing on in situ mechanically strained samples [332]. However, these techniques often inaccurately measure the extent of healing, as observed in various studies, leading to significant differences compared to the actual healing efficiency seen in mechanical tests, mainly due to limited surface penetration [333, 334]. Another concern when using light-based techniques to characterize SH polymer composites incorporating 2D materials pertains to the full spectrum absorption capacity exhibited by materials such as graphene [335]. This capability raises a potential issue wherein these materials could induce healing during testing, attributable to the photothermal effect initiated by light.

In a particular study, a pioneering method was adopted to investigate the SH mechanism, confirming it as radical-mediated disulfide exchange in graphene–vitrimer SH composites, using electron paramagnetic resonance (EPR) spectroscopy [336]. Emerging spectroscopy techniques such as solid-state nuclear magnetic resonance (SSNMR) [337], including variations like correlation spectroscopy (COSY) [338], nuclear Overhauser effect spectroscopy (NOESY) [338], and magic angle spinning SSNMR (MAS-SSNMR) [339], broadband dielectric spectroscopy (BDS) [340], and positron annihilation spectroscopy (PAS) [341] offer the potential to uncover microscale chemical interactions of composites. These techniques can elucidate aspects such as polymer chain flexibility, changes occurring at microsecond-to-picosecond timescales, properties of dangling chain ends, and correlations between chain properties and macrostructure. Despite their capabilities, these techniques have yet to be explored extensively for composites incorporating 2D materials. This unexplored territory leaves ample room for further investigation, particularly in understanding the influence of 2D materials on polymer chain dynamics.

A vast array of SH nanocomposites has been developed for applications in protective coatings, where 2D materials play a pivotal role in enhancing barrier properties in addition to strength and healing capabilities [214, 218, 342]. Consequently, specialized electrochemical techniques have been devised to understand the coating state by observing alterations in surface conductivity. One of the simplest techniques involves electrochemical impedance spectroscopy (EIS), wherein the bulk resistance of the entire exposed surface is measured by applying a sinusoidal perturbation [343]. Employing circuit fitting enables the estimation of individual circuit components, thereby identifying the coating's contribution. A more sophisticated approach of using scanning vibrating electrode technique (SVET) analysis is demonstrated in a study by Hao et al. [344]. This technique monitors current density at localized defects on the coating's surface. Defects exposing metal directly to the electrolyte exhibit distinct cathodic and anodic activity. As healing progresses, the current density decreases, augmenting surface resistance and diminishing cathodic and anodic activities. Presenting results in the form of a current density map facilitates the monitoring of progress across the crack geometry. However, a limitation of this technique lies in its reliance solely on resistance measurements. This resistance may also stem from the formation of an oxide layer on the surface, potentially skewing the map without actual healing taking place. Moreover, because this can measure conductivity with high precision, there is a possibility of hotspots caused by a localized decrease in resistivity due to improper dispersion of conductive 2D materials. Figure 13 summarizes the key measurements and material characterization techniques, along with their applications. These include understanding composite properties, measuring healing efficiency, detecting 2D fillers, identifying healing mechanisms, and analyzing 2D filler–matrix interactions.

Along with characterization techniques, it is very important to develop monitoring techniques where conditional monitoring of the SH process can be done to observe and track real-time changes in a material's properties, behavior, or structural integrity during the healing process. Among them, non-destructive testing (NDT) techniques are pivotal for identifying meso- and macroscopic damages in composite materials, with one highly effective method being the utilization of broad-band ultrasound [345]. Elastic waves with frequencies above 20 kHz serve as an excellent tool for damage detection. Through a meticulous analysis of ultrasonic wave reflection or transmission, various types of damage, including voids, cracks, and delamination, can be both detected and precisely localized [346]. Multiple variations have emerged for ultrasonic testing, such as air-coupled C-scan, water-coupled C-scan, and local defect resonance measurements, which are selected based on the material properties, structural assessment, and crack morphology [347]. For example, a comparative analysis was conducted on carbon fiber reinforced polymer (CFRP) composites featuring an ionomer interlayer [348]. This study involved the deliberate creation of artificial delamination of varying sizes and positions within the material during its production phase. The dimensions of these defects were assessed both before and after thermal healing using air-coupled C-scan, water-coupled C-scan, and local defect resonance measurements. Although these methods possess significant potential for monitoring, they have not been extensively investigated for SH polymeric nanocomposites.

The advanced characterization techniques previously discussed provide either qualitative or quantitative insights into the healing process. When quantified, this information is typically expressed as healing efficiency (%) derived from the ratio of property values after healing to those before healing. As emphasized, mechanical testing stands as the most dependable method for quantifying healing phenomena because the material often experiences structural damage, and the recovery in mechanical strength is of primary concern. However, in scenarios where the material is destined for diverse applications beyond regular mechanical stress, such as its role in EMI shielding, it becomes pertinent to gauge healing in alignment with those specific performance metrics. Consequently, healing efficiency can be determined as a function of multiple performance indicators, not solely limited to mechanical strength, but depending on the material's designated function. Previous studies have exemplified the use of various parameters, including tensile strength [154], elongation at break [349], flexural stiffness [350], electrical conductivity [351], EMI shielding efficacy [199], thermal conductivity [352], sensor performance [353], fracture toughness [324], crack dimensions [354], capacitance retention [207], and frequency response [355] to quantify the healing process. Although this diversity in parameters might complicate direct material comparisons across different applications, it significantly aids in comprehending the material's suitability and potential for specific applications.

## 6 | Future Outlook and Conclusion

Inspired by nature, the ability of engineered materials to self-heal is highly sought after and holds immense potential to transform our perception of man-made substances. Decades of research have been dedicated to finding suitable SH materials, aiming to

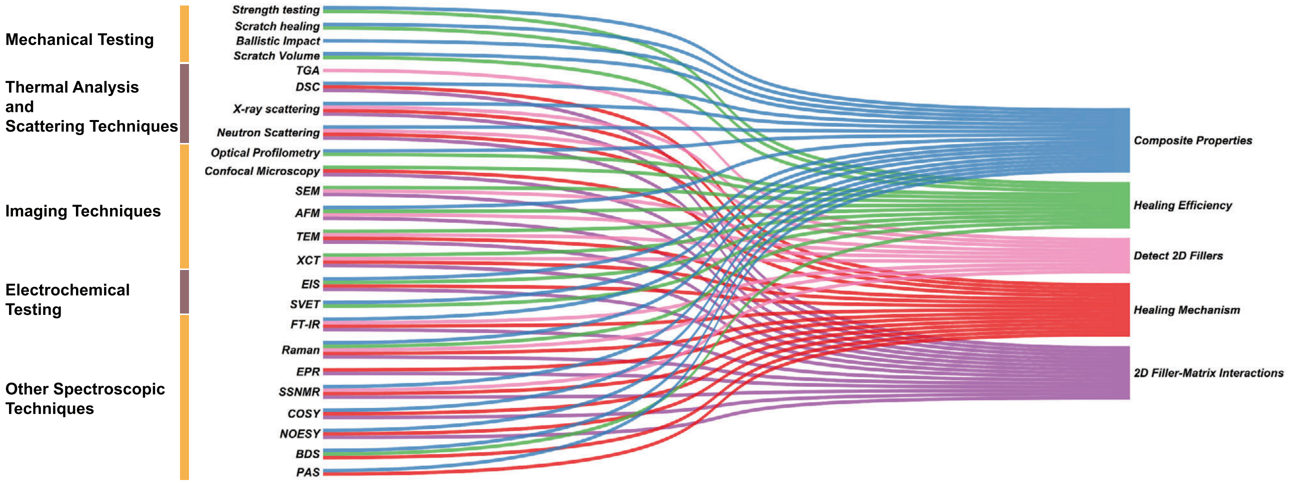

**FIGURE 13** | Summary of characterization techniques for SH polymer composites with 2D materials. AFM, atomic force microscope; BDS, broadband dielectric spectroscopy; COSY, correlation spectroscopy; EIS, electrochemical impedance spectroscopy; EPR, electron paramagnetic resonance; FT-IR, Fourier transform infrared spectroscopy; NOESY, nuclear Overhauser effect spectroscopy; PAS, positron annihilation spectroscopy; SEM, scanning electron microscope; SSNMR, solid-state nuclear magnetic resonance; SVET, scanning vibrating electrode technique; TEM, transmission electron microscope.

create durable and repairable materials with exceptional healing abilities and consistency. Many supramolecular structures have been adapted or created in pursuit of this goal. However, before these products can be widely adopted commercially, certain challenges must be addressed. For instance, achieving high healing efficiency within the polymer matrix requires using highly dynamic networks with significant polymer chain mobility. Yet, this pursuit often leads to a compromise in mechanical strength. To counter this strength loss, the addition of reinforcing fillers like 2D materials is one of the suitable approaches.

Incorporating a specific amount of 2D materials significantly boosts the mechanical strength of the composite, which holds great promise in using SH polymeric materials for lightweight aerospace components. The added functionality from materials like graphene brings improved electrical conductivity, which, in turn, could enhance resistance against lightning strikes, for example. Moreover, it opens up possibilities for new ways to trigger healing in the composite, such as through joule heating while also using this energy for de-icing. In certain scenarios, the stimulus required for healing is readily available at the application site. For instance, in the case of using these composites for battery pack casings, heat stimulus is easily accessible. This available energy can be harnessed to not only heal the structures but also efficiently dissipate heat, offering a dual advantage in both repair and thermal management. Similarly, in situ polymerization processes that require metal catalysts for successful network formation can use metal particles grown on atomically thin materials, which can help in polymerization while also reinforcing the matrix [356]. This capability aligns with potential applications in various industries, promising innovative solutions for structural maintenance and performance enhancement.

However, in engineering applications, it is not always feasible to trigger healing solely through external stimuli. Repairing mechanically robust polymers becomes challenging when there's a lack of stimulus, as the polymer chains remain in a "frozen" state below the glass-transition temperature of the host matrix [357]. This difficulty is compounded when using 2D materials for reinforcement, as they tend to further limit chain mobility by creating structural barriers at the nano and micro levels. A potential solution lies in utilizing these 2D materials not only for mechanical reinforcement but also integrating them chemically into the healing process through surface functionalization. By introducing functional groups on the surface, the adhesion between the 2D fillers and matrix can be enhanced, enabling the formation of reversible bonds during the healing process, depending on the introduced functional group. Despite its seeming simplicity, the ease and extent of functionalization heavily rely on various factors like the physical morphology of the materials, sheet defects, synthesis method, and even the solvent used for exfoliation or functionalization. Hence, to ensure effective bonding between the nanosheets and the host matrix, meticulous attention is necessary throughout the process, from synthesis to incorporation into the matrix. Achieving control over this bonding chemistry holds the key to unlocking the full potential of these materials for SH applications in real-world engineering scenarios.

When it comes to the physical morphology of the nanosheets, factors like lateral size and layer count of the 2D materials highly influence the performance of the composites. In a study by Lin et al., a simple DA-PU matrix was used to test the effect of lateral size and effect of functionalization on GO nanosheets, and it was found that laterally small nanosheets result in mechanically strong composites with higher healing efficiency [358]. This enhancement can be directly ascribed to the reduced hindrance in the mobility of polymer chains due to the presence of smaller nanosheets within the matrix. Furthermore, the study showcased that functionalizing nanosheets with maleimide moieties contributed to an additional augmentation in both the strength and healing efficiency of the composite materials. These findings underscore the pivotal role of nanosheet characteristics and surface modifications in optimizing composite performance. In situations where preserving the physical characteristics of the polymer is crucial, and yet reaping the benefits of 2D materials like enhanced performance in specific applications (such as sensing materials) is desired without their direct integration into the matrix, less invasive methods like the layer-by-layer construction technique can be employed [246]. This approach allows leveraging the advantages of both materials without altering the inherent properties of the polymer.

Figure 14A,B illustrates the healing efficiency and mechanical strength of SH polymer composites reinforced with various 2D materials. In this statistical analysis shown in Figure 14C, certain studies report multiple healing efficiencies due to fluctuations in nanofiller loading and healing stimuli. Hence, the highest reported values are considered for comparison. Interestingly, most of the reported nanocomposites demonstrate relatively higher healing efficiency (signified by high recovery zone), with many approaching near-perfect scores close to 100%. Initially hypothesized correlations between factors such as nanosheet type, surface functionalization, or matrix type; however, revealed a lack of discernible trends in the obtained data. This suggests that numerous other variables, including nanosheet properties (e.g., lateral size distribution, layer count distribution, and surface defects), matrix properties (e.g., molecular weight, crystallinity, and Tg), and interface properties (e.g., cut sharpness and applied force during reconnection), significantly influence the overall system properties. Despite graphene exhibiting nearly 10-fold higher tensile strength compared to some other 2D materials, this dominance in strength does not consistently manifest in the experimental data [53]. This could be because certain composites displaying exceptionally high strength were found to be reinforced with additional materials like glass or aramid fibers. To confirm this observation, a comprehensive case study focusing on PU was conducted as shown in Figure 15 where only PU and 2D material composites were considered. The examination of studies utilizing PU as the matrix aimed to analyze the impact of nanosheet type and surface functionalization on both tensile strength and healing efficiency. However, similar unpredictability in the data emerged, underscoring the necessity for further studies on the influence of other parameters and standardized evaluation practices in assessing healing properties. Identifying fundamental factors amid this complexity is crucial for future advancements in this field.

The extensive use of solvents in handling 2D materials raises environmental concerns. Lee et al. introduced a solvent-free approach using a one-shot process, bonding GNP through DA interactions while simultaneously polymerizing PU [359]. This method eliminates the need for solvent-based steps in nanocomposite synthesis, offering a more eco-friendly approach. More innovations in this arena are needed to mitigate the impact of hazardous solvents and nanowaste on the environment [360].

In numerous methods aimed at healing composite materials, a crucial step involves manually aligning or bringing together the fractured components before the chemical mending mechanisms can initiate. This manual alignment poses a significant challenge, especially in practical engineering applications where relatively large structures cannot easily be reconnected by simple physical manipulation. Attempting to do so might even exacerbate the damage incurred. To address this challenge, novel technologies such as SMASH and close-then-heal (CTH) have been under exploration [361]. Most of the other techniques share a common prerequisite: The fractured parts must be positioned in contact before the healing process commences. From these, CTH presents promise for real-world applications due to its reliance on constrained shape recovery. This means that the shape memory effect will help position the damaged ends of the composite together, whereas the healing chemistry will play its role in reconnecting thereafter. This aspect aligns well with the constraints often observed in load-bearing structures, which are typically fixed or supported at their edges and are subjected to external forces, like tension or compression, during the healing phase. Although the integration of these healing techniques with 2D materials has not been extensively investigated, it would be interesting to see the effect on healing in nanocomposites as at least graphene out of all 2D materials is known to endorse the shape memory actuation [362].

The addition of 2D materials introduces a spectrum of supplementary interactions that necessitate thorough investigation to comprehend the system's state. This scrutiny begins before integrating the 2D materials into the host matrix, where an assessment of their physical and chemical properties is conducted. Certain properties can be evaluated pre-incorporation into the polymer matrix, providing a reliable insight into the material's suitability for specific applications. For instance, employing solid-state ultraviolet–visible–NIR (UV–Vis–NIR) spectroscopy and EPR enables the examination of the light absorbance properties of these nanomaterials [390]. This evaluation serves as a pivotal indicator for assessing the compatibility of the 2D materials within the overall matrix. Furthermore, it offers valuable insights into the potential efficacy of these 2D materials, specifically in enhancing the photo-healing performance of the composite matrix. Additional monitoring techniques, such as Raman spectroscopy, aid in comprehending critical aspects like stress distribution and load transfer occurring at the interface between the 2D material and the polymer. Moreover, these techniques facilitate sophisticated modeling employing continuum mechanics principles. This allows for a more comprehensive understanding of the structural behavior and mechanical interactions within the composite system, enhancing our ability to optimize the performance of 2D material-based SH composites.

The molecular level rebonding interactions crucial for polymer synthesis and healing can be significantly influenced by the presence of moisture or oxygen, particularly impacting these processes at ambient temperatures. Integrating laterally large 2D materials into the polymer matrix creates a convoluted path, effectively reducing moisture seepage into the matrix. These 2D nanosheets offer an intriguing capability: They can be intentionally aligned within the polymer to create microchannels [391]. This alignment serves to confine the movement of polymer chains, especially in the anticipated direction of damage. Consequently, when a stimulus is applied to the material as a whole, the movement of chains aligns favorably with the direction of these oriented nanosheets, thereby optimizing the healing response.

The utilization of multiple 2D materials in SH polymers is less prevalent than anticipated due to their comparable properties and the limited potential for synergistic effects. The similarity in morphology-dependent properties with respect to mechanical

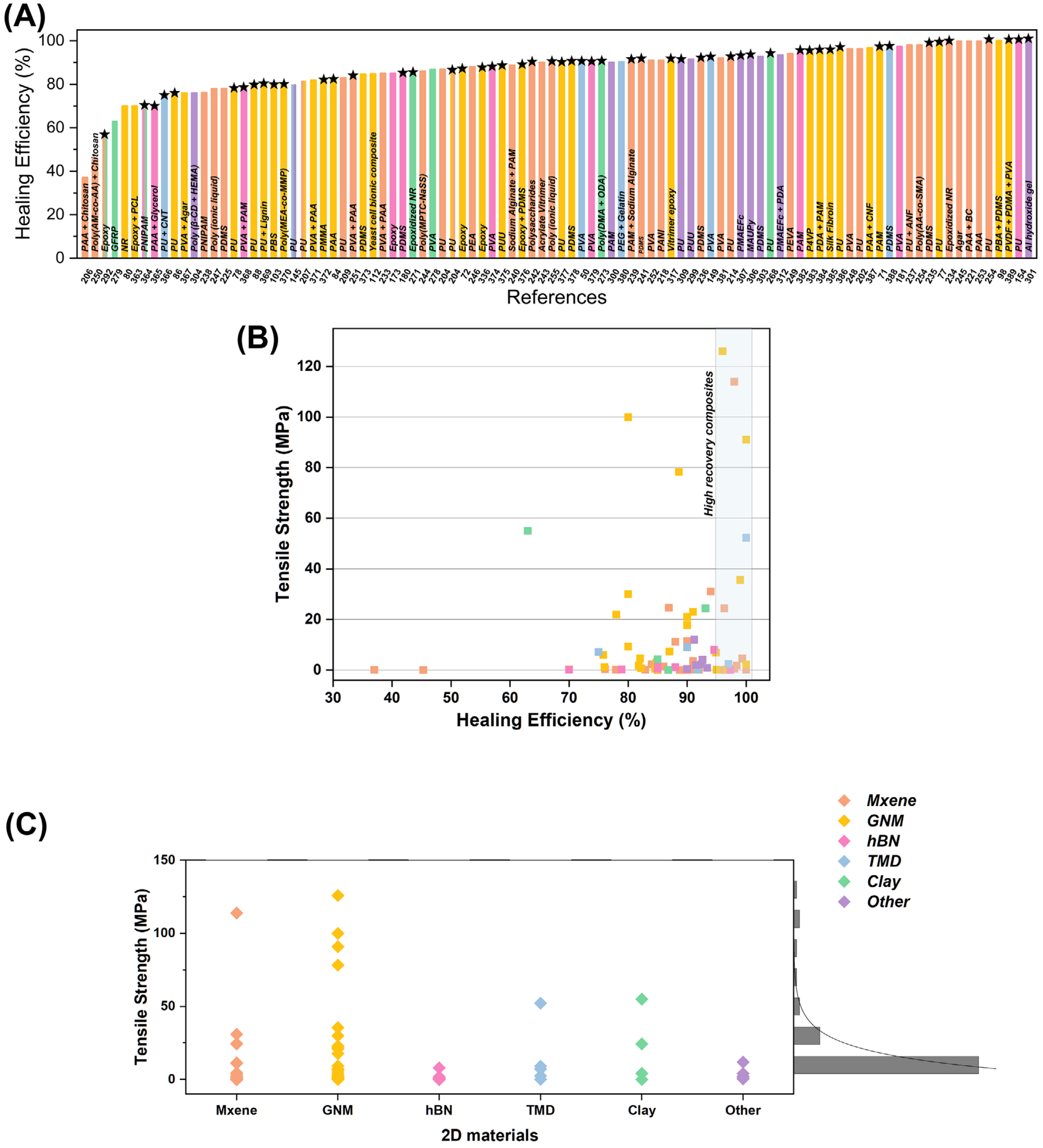

**FIGURE 14** | Plots from surveyed literature of (A) healing efficiency, (B) tensile strength, and (C) statistical analysis of tensile strength of the self-healable composites made from 2D materials. ★ means that the 2D materials used in the composite have been functionalized. Colors of the bars and scatter points represent the type of 2D material used, and the legend is shown in (C). Values written vertically on the bar show the polymer matrix used in that study. ANF, aramid nanofibers; GFRP, glass fiber reinforced plastic; MAUPy, 2-(3-(6-methyl-4-oxo-1,2,3,4-tetrahydropyrimidin-2-yl)ureido)ethyl methacrylate; NR, natural rubber; P4VP, poly(4-vinylpyridine); PAA, polyacrylic acid; PAM, polyacrylamide; PANI, polyaniline; PBS, polyborosiloxane; PDMS, polydimethylsiloxane; PEG, polyethylene glycol; PEVA, poly(ethylene-vinyl acetate); PMAEFc, poly(2-aryloyloxy)ether ferrocenecarboxylate; PMMA, poly(methyl methacrylate); PNIPAM, poly(*N*-isopropylacrylamide); PU, polyurethane; PUU, poly(urea–urethane); PVA, polyvinyl alcohol; TMD, transition metal dichalcogenide.

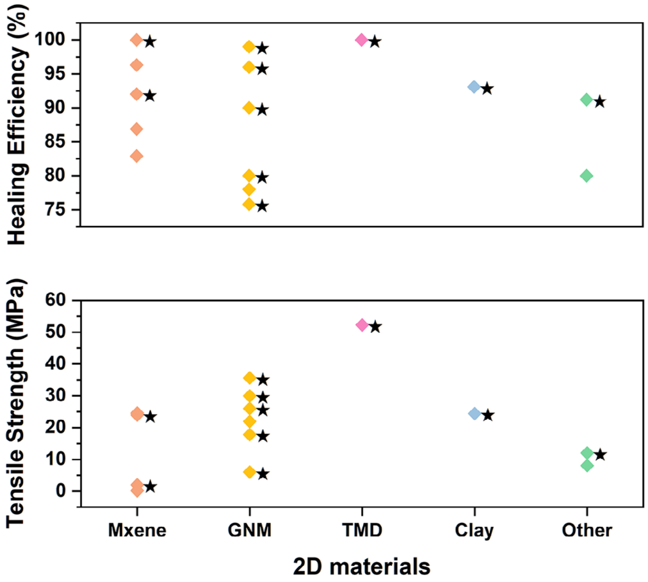

**FIGURE 15** | Comparative analysis of PU reinforced with different 2D materials obtained from the literature. TMD, transition metal dichalcogenide.

reinforcement restricts the opportunities for achieving synergistic benefits in strength, diminishing the incentive for incorporating multiple components at the cost of system complexity. For future works, rather than solely focusing on permutations of nanosheet sources, nanosheet exfoliation techniques, or host polymers, it is imperative to shift attention towards comprehending the fundamental aspects of the physical and chemical healing processes at the molecular level. This deeper understanding can enable the precise control and customization of interactions, catering to specific requirements. Developing robust theoretical models around these healing phenomena holds promise for challenging existing engineering materials, which are more economically scaled and readily available. Realizing this prospect necessitates standardized testing methodologies across systems and a deeper investigation into the intricacies of healing interfacial processes.

In the pursuit of advancing SH polymers, the overarching goal lies in identifying materials that emulate organism-like material capabilities. These materials should demonstrate the capacity to detect structural damage, react to environmental stimuli, and instigate adaptive responses. Leveraging the remarkable multifunctional attributes of 2D materials is critical in the construction of intricately designed smart devices and systems. Within these systems, the objective of healing transcends conventional mechanical restoration; it involves the automated detection of damage and the initiation of autonomous repairs, integrating time-sensitive properties into the healing process.

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## Conflicts of Interest

The authors declare no conflicts of interest.

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