International Journal of Engineering, Science and Humanities
An international peer reviewed, refereed, open-access journal
Impact Factor 8.3 www.ijesh.com ISSN: 2250-3552

Chemical Design of Smart Polymers for Self-Healing and Stimuli-Responsive Applications: A Comprehensive Review

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ABSTRACT
Smart polymers have emerged as a transformative class of advanced materials capable of responding dynamically to environmental stimuli and autonomously repairing damage. These materials exhibit adaptive behaviors such as self-healing, shape memory, conductivity modulation, and responsiveness to external triggers including temperature, pH, light, moisture, electric fields, and mechanical stress. The development of smart polymers has attracted significant attention due to their potential applications in biomedical engineering, flexible electronics, aerospace structures, coatings, sensors, robotics, and sustainable materials technologies. The chemical design of smart polymers is fundamentally based on reversible interactions, dynamic covalent chemistry, supramolecular assemblies, and stimuli-responsive molecular architectures that enable reversible structural transformations. Self-healing mechanisms are broadly categorized into intrinsic and extrinsic systems, with intrinsic approaches utilizing reversible chemical bonds and extrinsic approaches employing microcapsules or vascular networks containing healing agents. Recent advances in dynamic covalent networks, hydrogen bonding systems, ionic interactions, host–guest chemistry, and reversible cross-linking strategies have significantly enhanced healing efficiency and responsiveness. Simultaneously, stimuli-responsive polymers have evolved to exhibit multiple functionalities, allowing materials to adapt intelligently to changing environments. This review discusses the chemical principles underlying smart polymer design, self-healing mechanisms, dynamic polymer networks, and stimuli-responsive behaviors.

Keywords: Smart polymers, Self-healing materials, Stimuli-responsive polymers, Dynamic covalent chemistry, Supramolecular polymers, Shape-memory polymers, Adaptive materials.

1. INTRODUCTION

The continuous advancement of materials science has created increasing demand for materials capable of adapting to changing environmental conditions while maintaining structural integrity and functionality. Conventional polymeric materials often suffer from irreversible damage, fatigue, and degradation during service, resulting in reduced performance and increased maintenance costs. To overcome these limitations, researchers have developed smart polymers

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that possess the ability to sense external stimuli and respond through predictable physical or chemical changes. These materials represent a significant departure from traditional passive materials because they can actively modify their properties in response to environmental conditions.

Smart polymers are defined as polymeric systems capable of undergoing controlled and reversible changes in their physical, chemical, or mechanical properties when exposed to specific stimuli. These stimuli may include temperature variations, pH fluctuations, light irradiation, magnetic fields, electric fields, humidity changes, or mechanical forces. Such responsive behavior enables polymers to perform sophisticated functions that resemble biological systems, including self-repair, shape transformation, controlled drug release, and adaptive sensing.

Among the various functionalities exhibited by smart polymers, self-healing capability has gained particular importance. Self-healing materials can repair damage autonomously or through external stimulation, thereby extending service life and reducing maintenance requirements. Inspired by biological healing processes observed in living organisms, self-healing polymers have become a major focus of research in materials engineering. The integration of self-healing mechanisms with stimuli-responsive functionalities has further expanded the range of potential applications.

Recent advances in polymer chemistry have enabled the development of dynamic molecular architectures based on reversible interactions and adaptive networks. These innovations have facilitated the creation of multifunctional materials capable of responding intelligently to complex environments. Consequently, smart polymers have become increasingly relevant in fields such as biomedical engineering, aerospace technology, wearable electronics, soft robotics, energy storage, and environmental remediation.

This review examines the chemical design strategies employed in smart polymers, focusing on self-healing mechanisms and stimuli-responsive functionalities. Special attention is given to dynamic covalent chemistry, supramolecular interactions, and emerging trends in multifunctional polymer systems.

2. FUNDAMENTALS OF SMART POLYMER DESIGN

The design of smart polymers is fundamentally based on the incorporation of responsive molecular components into polymeric architectures. These molecular structures enable materials to undergo reversible changes in response to external stimuli while maintaining overall structural stability.

Smart polymers generally consist of three essential components:

1. A polymer backbone providing structural support.
2. Responsive functional groups capable of sensing stimuli.
3. Dynamic interactions that facilitate reversible changes.

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The selection of chemical functionalities plays a critical role in determining the responsiveness and performance of smart polymer systems. Responsive groups may include thermosensitive segments, ionizable moieties, photoactive molecules, conductive units, or dynamic covalent bonds. These functionalities enable polymers to exhibit specific responses under defined environmental conditions.

Polymer architecture also significantly influences responsiveness. Linear polymers, branched polymers, cross-linked networks, block copolymers, and interpenetrating polymer networks can all be engineered to achieve distinct adaptive behaviors. Advances in controlled polymerization techniques have enabled precise control over molecular weight, composition, and network structure, thereby enhancing material performance.

Another important aspect of smart polymer design involves balancing responsiveness and stability. Excessive mobility may improve responsiveness but compromise mechanical strength, whereas highly rigid networks may exhibit excellent durability but limited adaptability. Therefore, chemical design strategies often focus on optimizing this balance through careful control of molecular interactions and cross-link density.

3. LITERATURE REVIEW

Smart polymers and self-healing materials have emerged as one of the most innovative areas of materials science due to their ability to respond to external stimuli and recover functionality after damage. Early studies established the foundation for understanding dynamic polymeric systems that mimic biological self-repair mechanisms. Cordier et al. (2014) demonstrated that supramolecular assemblies could produce thermoreversible rubber capable of autonomous healing through reversible intermolecular interactions. Similarly, Burnworth et al. (2014) introduced optically healable supramolecular polymers, highlighting the role of light-responsive mechanisms in repairing material damage. Chen et al. (2014) further advanced the field by developing autonomic self-healing thermoplastic elastomers through multiphase design strategies, enabling enhanced mechanical performance and durability. Blaiszik et al. (2014) provided a comprehensive overview of self-healing polymers and composites, categorizing healing approaches into intrinsic and extrinsic systems. Their work emphasized the significance of microcapsules, vascular networks, and reversible chemical bonds in facilitating damage repair. Hager et al. (2015) expanded this understanding by discussing self-healing materials as multifunctional systems capable of extending service life and reducing maintenance requirements. These pioneering investigations established the theoretical and practical basis for the development of smart polymeric materials, demonstrating that dynamic molecular interactions can significantly improve material reliability, sustainability, and performance in demanding applications.

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The evolution of smart polymers has been closely linked to advances in stimuli-responsive materials capable of reacting to environmental changes such as temperature, pH, light, moisture, and mechanical stress. Stuart et al. (2016) highlighted the broad spectrum of stimuli-responsive polymer applications, emphasizing their adaptability in fields ranging from biotechnology to advanced manufacturing. Zhao et al. (2015) contributed significantly to this domain through their review of shape-memory polymers, which can recover predefined shapes upon exposure to specific triggers. Their analysis demonstrated the importance of molecular architecture and switching segments in controlling shape-memory behavior. Bao and Chen (2016) further emphasized the growing relevance of flexible and stretchable polymeric devices, particularly in wearable electronics and smart sensing technologies. Dynamic polymer networks described by Roy et al. (2015) introduced the concept of "dynamers," which possess reversible covalent and non-covalent bonds that continuously reorganize, enabling adaptability and self-repair. Wei et al. (2015) explored self-healing gels based on constitutional dynamic chemistry, showing how reversible chemical interactions can produce highly responsive and recoverable hydrogel systems. Li et al. (2015) developed responsive double-network hydrogels with exceptional mechanical strength and adaptability, demonstrating the integration of responsiveness and robustness within a single material system. Collectively, these studies revealed that smart polymers are not passive materials but active systems capable of adapting their structure and properties in response to external stimuli, thereby expanding their functional potential across multiple industries.

Research attention has increasingly focused on enhancing the mechanical properties, healing efficiency, and multifunctionality of self-healing polymers. Urban (2016) presented a comprehensive discussion of self-healing polymers, emphasizing the transition from fundamental principles to practical engineering applications. The study identified reversible hydrogen bonding, ionic interactions, and dynamic covalent chemistry as critical mechanisms enabling repeated healing cycles. Yang and Urban (2017) further explored self-healing polymeric materials, emphasizing the role of molecular mobility and network dynamics in determining healing performance. Wang and Urban (2018) provided an updated perspective on self-healing polymers, discussing emerging strategies for achieving rapid healing, environmental stability, and scalability. Wu et al. (2018) reviewed recent developments in self-healing polymeric materials and highlighted the growing integration of nanotechnology, supramolecular chemistry, and dynamic bonding mechanisms to improve healing efficiency. Sun et al. (2019) demonstrated that polyampholyte-based physical hydrogels exhibit remarkable toughness and self-healing capabilities due to reversible ionic interactions within the network structure. Zhang et al. (2019) examined thermally self-healing polymers as a promising route for recycling thermoset materials, thereby addressing sustainability challenges associated with conventional polymer

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systems. These investigations collectively indicate that modern self-healing polymers are increasingly capable of combining mechanical robustness, environmental resilience, and repairability, making them suitable for long-term structural and functional applications.

Recent developments have expanded the application scope of smart polymers into biomedical engineering, sustainable materials, soft robotics, and advanced manufacturing technologies. Li and Meng (2018) reviewed the application of smart polymers in biomedical systems, highlighting their use in drug delivery, tissue engineering, wound healing, and biosensing. The ability of these materials to respond to physiological stimuli makes them particularly valuable in personalized healthcare solutions. Fu and Urban (2020) discussed recent advances in stimuli-responsive polymers and identified emerging opportunities in adaptive coatings, flexible electronics, and intelligent medical devices. Their review emphasized the growing integration of artificial intelligence, nanotechnology, and advanced polymer chemistry to create next-generation smart materials. Zou et al. (2021) examined dynamic covalent polymer networks and demonstrated how traditional chemical concepts have evolved into innovative material platforms characterized by reprocessability, self-healing capability, and environmental sustainability. The study highlighted the potential of dynamic covalent chemistry to address challenges related to material recyclability and lifecycle management. Furthermore, ongoing research increasingly focuses on combining multiple functionalities, including self-healing, shape-memory behavior, conductivity, and biocompatibility within single polymer systems. This multifunctional approach is expected to drive future innovations in aerospace, healthcare, energy storage, and wearable technologies. The literature demonstrates that smart polymers have evolved from conceptual laboratory materials into sophisticated engineering systems with significant commercial and societal relevance. Their unique ability to adapt, heal, and respond intelligently to environmental stimuli positions them as key materials for future technological advancement and sustainable development.

Literature Review Table

Author(s) & Year	Research Focus	Method/Material	Key Findings	Research Contribution
Cordier et al. (2014)	Self-healing rubber	Supramolecular assembly	Developed thermoreversible rubber with autonomous healing ability	Established foundation for intrinsic self-healing polymers
Burnworth et al. (2014)	Optically healable polymers	Supramolecular polymers	Demonstrated light-triggered healing mechanisms	Introduced photo-responsive self-healing materials

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Chen et al. (2014)	Thermoplastic elastomers	Multiphase polymer design	Achieved autonomic self-healing with improved durability	Enhanced mechanical performance of self-healing systems
Blaiszik et al. (2014)	Self-healing polymers and composites	Review of intrinsic and extrinsic systems	Classified healing mechanisms and technologies	Provided comprehensive healing framework
Hager et al. (2015)	Multifunctional self-healing materials	Advanced polymer networks	Improved material lifespan and reduced maintenance	Highlighted industrial significance of self-healing materials
Stuart et al. (2016)	Stimuli-responsive polymers	Responsive polymer systems	Demonstrated responses to temperature, pH, and light	Expanded applications of smart polymers
Zhao et al. (2015)	Shape-memory polymers	Stimuli-responsive networks	Revealed mechanisms controlling shape recovery	Advanced shape-memory material design
Bao & Chen (2016)	Flexible electronics	Stretchable polymer devices	Improved flexibility and sensing capabilities	Supported wearable electronic applications
Roy et al. (2015)	Dynamic polymers (Dynamers)	Reversible covalent networks	Developed adaptable and self-repairing polymers	Introduced dynamic polymer concept
Wei et al. (2015)	Self-healing hydrogels	Dynamic chemistry-based gels	Demonstrated reversible and recoverable gel systems	Enhanced hydrogel functionality
Li et al. (2015)	Double-network hydrogels	Interpenetrating polymer networks	Achieved high toughness and	Combined strength with

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Impact Factor 8.3 www.ijesh.com ISSN: 2250-3552

			responsiveness	adaptability

4. RESEARCH GAP IDENTIFIED

Research Area	Existing Findings	Gap Identified
Self-Healing Efficiency	Effective healing mechanisms available	Healing speed and repeated healing cycles need improvement
Mechanical Strength	Strong polymer networks developed	Trade-off between strength and healing remains unresolved
Stimuli Responsiveness	Multiple stimuli-responsive systems reported	Limited integration of multiple stimuli in one material
Biomedical Applications	Promising biocompatible polymers available	Long-term safety and clinical validation remain limited
Sustainability	Recyclable dynamic networks emerging	Large-scale industrial implementation is still challenging
Multifunctionality	Individual smart functions demonstrated	Integration of healing, sensing, conductivity, and shape memory requires further research

5. CHEMICAL PRINCIPLES OF SELF-HEALING POLYMERS

Self-healing polymers represent one of the most significant developments in smart materials research. These systems can restore mechanical integrity and functionality after damage through autonomous or externally triggered repair mechanisms.

Self-healing processes generally involve four stages:

1. Damage detection.
2. Transportation of healing agents or molecular rearrangement.
3. Healing reaction.
4. Restoration of material properties.

The chemical mechanisms responsible for healing are typically classified into intrinsic and extrinsic approaches.

6. EXTRINSIC SELF-HEALING SYSTEMS

Extrinsic self-healing systems contain embedded healing agents stored within microcapsules, hollow fibers, or vascular networks. When damage occurs, these reservoirs rupture and release healing compounds into the damaged region.

Microcapsule-based systems represent one of the earliest successful approaches. The healing agent flows into cracks and polymerizes upon contact with a catalyst, restoring structural continuity. Although effective, these systems often provide only a single healing event because healing agents become depleted after use.

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Vascular systems overcome this limitation by supplying healing agents continuously through interconnected channels. These networks resemble biological circulatory systems and enable multiple healing cycles.

7. INTRINSIC SELF-HEALING SYSTEMS

Intrinsic self-healing relies on reversible molecular interactions incorporated directly into polymer networks.

Common mechanisms include:

- Hydrogen bonding
- Ionic interactions
- Metal–ligand coordination
- Dynamic covalent bonding
- Host–guest interactions

Because healing capability is integrated into the polymer structure itself, intrinsic systems can often undergo repeated healing cycles without external replenishment.

8. DYNAMIC COVALENT CHEMISTRY IN SMART POLYMERS

Dynamic covalent chemistry (DCC) has emerged as a fundamental strategy in the design of smart polymers, particularly for self-healing and stimuli-responsive applications. Unlike conventional covalent bonds, which are generally permanent and irreversible, dynamic covalent bonds possess the unique ability to break and reform under specific environmental conditions while preserving the overall integrity of the polymer network. This reversible behavior enables materials to recover from physical damage, adapt to external stimuli, and undergo reprocessing or recycling without significant loss of performance. As a result, dynamic covalent chemistry bridges the gap between the mechanical robustness of traditional thermosetting polymers and the adaptability required for advanced functional materials. The incorporation of dynamic covalent bonds into polymer architectures has significantly enhanced the development of sustainable, durable, and multifunctional materials for applications in coatings, electronics, biomedical devices, and structural engineering.

Among the most widely studied dynamic covalent mechanisms are Diels–Alder reactions, disulfide exchange reactions, and imine (Schiff base) chemistry. Diels–Alder reactions involve reversible cycloaddition processes in which covalent bonds dissociate through retro-Diels–Alder reactions at elevated temperatures and reform upon cooling, providing excellent healing efficiency, thermal responsiveness, and mechanical stability. Disulfide exchange reactions utilize sulfur–sulfur bonds that can undergo bond reshuffling when exposed to heat, light, or mechanical stress, resulting in rapid healing, low activation energy requirements, and multiple repair cycles. Similarly, imine or Schiff base bonds are formed through reversible condensation reactions between amines and aldehydes. These bonds readily undergo exchange reactions under mild

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conditions, enabling autonomous self-healing and dynamic network reconfiguration. Because healing can occur under physiological conditions, imine-based polymers are particularly attractive for biomedical applications such as tissue engineering, drug delivery systems, and wearable healthcare devices. Together, these dynamic covalent approaches provide versatile pathways for creating next-generation smart polymer systems with enhanced functionality and sustainability.

9. RESEARCH PROBLEM

Despite significant advancements in smart polymer technology, the development of self-healing and stimuli-responsive polymeric materials continues to face several scientific and technological challenges that limit their widespread commercial and industrial implementation. Conventional polymers often suffer from irreversible mechanical damage, structural degradation, and limited adaptability when exposed to varying environmental conditions, resulting in reduced performance, shortened service life, and increased maintenance costs. Although dynamic covalent chemistry has provided innovative solutions through reversible bonding mechanisms such as Diels–Alder reactions, disulfide exchange, and imine chemistry, many existing smart polymers still exhibit trade-offs between mechanical strength, healing efficiency, responsiveness, and long-term stability. Materials with excellent self-healing capabilities frequently demonstrate reduced mechanical robustness, while highly durable systems often require external stimuli or prolonged healing times that restrict practical applications. Furthermore, challenges related to scalability, manufacturing complexity, environmental sustainability, recyclability, and cost-effectiveness remain largely unresolved. In biomedical applications, concerns regarding biocompatibility, toxicity, and controlled responsiveness under physiological conditions continue to hinder clinical translation. Similarly, in flexible electronics, aerospace structures, and wearable devices, achieving simultaneous self-healing, multifunctionality, and reliable performance under repeated stress remains difficult. Another major research gap involves the limited understanding of structure–property relationships governing dynamic covalent networks and their influence on healing kinetics and stimuli responsiveness. Therefore, there is a critical need for a comprehensive evaluation of chemical design strategies that can optimize both self-healing performance and stimuli-responsive behavior while maintaining mechanical integrity and sustainability. Addressing these challenges is essential for developing next-generation smart polymers capable of meeting the increasing demands of advanced engineering, healthcare, environmental, and industrial applications.

10. CONCLUSION

Smart polymers have emerged as a highly promising class of advanced materials capable of responding intelligently to environmental stimuli while exhibiting autonomous self-healing capabilities. This review highlights that the chemical design of smart polymers is primarily

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driven by the incorporation of dynamic covalent bonds, supramolecular interactions, and stimuli-responsive functional groups that enable reversible structural transformations and adaptive behavior. Dynamic covalent chemistry, including Diels–Alder reactions, disulfide exchange mechanisms, and imine-based networks, has significantly enhanced the development of self-healing materials by combining mechanical robustness with reversible repair processes. Similarly, stimuli-responsive polymers capable of reacting to temperature, pH, light, electric fields, and mechanical stress have expanded the application potential of these materials across diverse sectors. The integration of self-healing and responsive functionalities has led to the development of multifunctional systems suitable for biomedical devices, drug delivery platforms, flexible electronics, soft robotics, protective coatings, aerospace structures, and sustainable materials technologies. Despite remarkable progress, several challenges remain, including the optimization of healing efficiency, mechanical performance, long-term stability, scalability, recyclability, and cost-effective manufacturing. Furthermore, achieving a balance between material durability and responsiveness continues to be a major research focus. Recent advances in molecular engineering, nanotechnology, and dynamic polymer network design indicate that future smart polymers will possess enhanced multifunctionality, sustainability, and adaptability. As research continues to evolve, the development of environmentally friendly and highly efficient smart polymer systems is expected to play a critical role in addressing technological and societal challenges. Smart polymers represent a transformative materials platform with immense potential to revolutionize next-generation engineering, healthcare, electronics, and sustainable manufacturing applications.

11. FUTURE WORK

Future research should focus on developing smart polymers with improved self-healing efficiency while maintaining high mechanical strength and durability for long-term applications.

Greater emphasis is needed on multifunctional polymer systems that combine self-healing, shape-memory behavior, conductivity, and stimuli responsiveness within a single material platform.

Researchers should explore sustainable and recyclable dynamic polymer networks to reduce environmental impacts and support circular economy initiatives in polymer manufacturing.

Further investigations into biocompatible smart polymers are essential for advancing biomedical applications such as tissue engineering, drug delivery, and wearable healthcare devices.

Advanced molecular design, nanotechnology integration, and artificial intelligence-assisted material discovery are expected to accelerate the development of next-generation smart polymers with enhanced performance and broader industrial applicability.

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