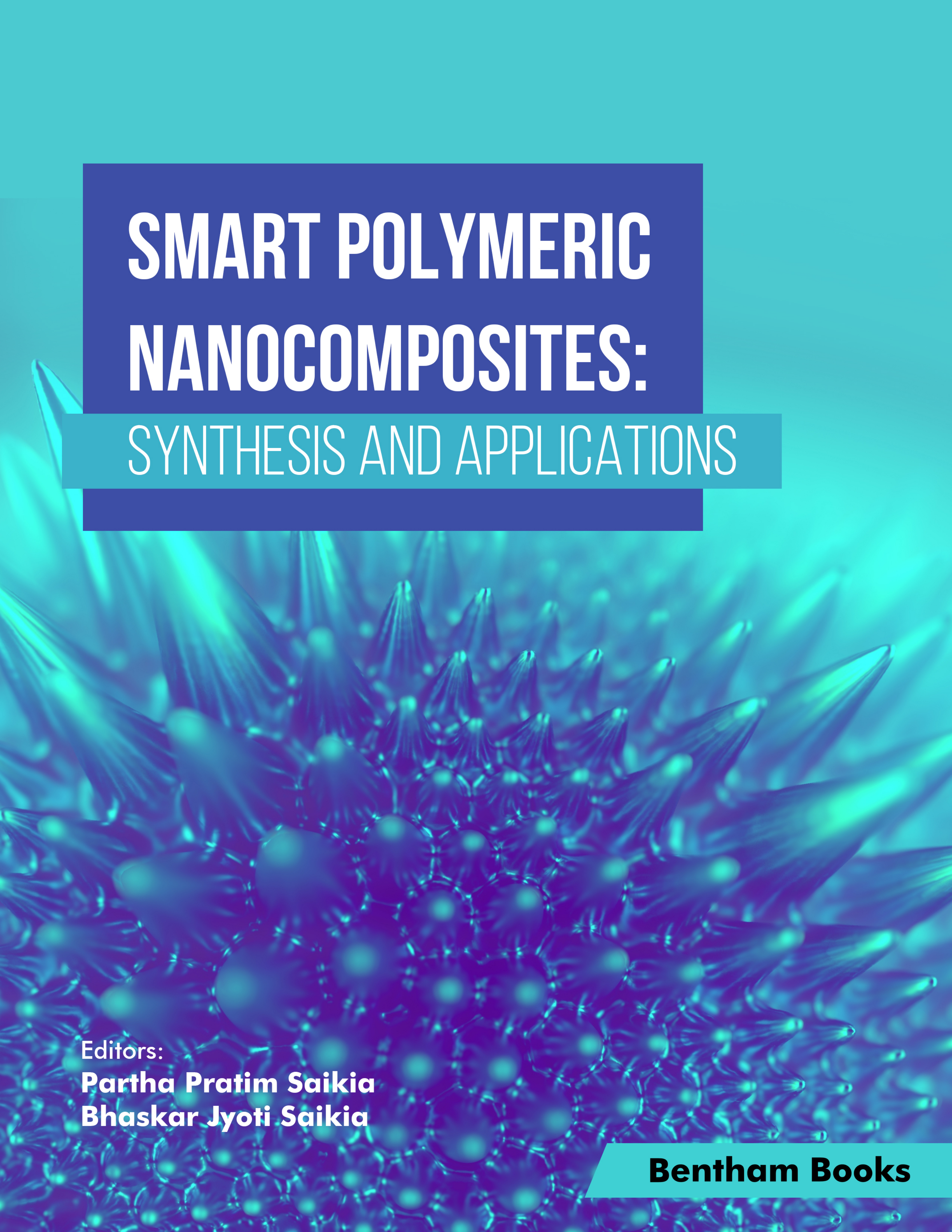
SMART POLYMERIC NANOCOMPOSITES:

SYNTHESIS AND APPLICATIONS



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Smart Polymeric Nanocomposites: Synthesis and Applications

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FOREWORD

Considering the importance of smart polymeric nanocomposites, this book covers a wide variety of chapters covering various aspects of this growing field. This is a huge asset for researchers particularly those who are working in the field of smart polymeric materials. Those who wish to initiate their research in this field can consult this book as a reference as it gives a glimpse of the latest developments in the field of nanocomposites. I hope this book will be beneficial for students of higher education as a part of their curriculum. Covering many aspects of smart polymeric nanocomposites starting from their synthesis, characterization, and applications in various fields such as self-healing, shape memory, drug delivery, water purification, flexible electronic sensing, *etc* it equips readers with a holistic understanding of smart polymeric nanocomposites.

With contributions from esteemed researchers and experts in the field, "**Smart Polymeric Nanocomposites: Synthesis and Applications**" offers a comprehensive overview of the latest advancements and future directions in this exciting area of study.

I hope this book serves as a valuable resource for researchers, academics, and industry professionals alike, inspiring further exploration and innovation in the field of smart polymeric nanocomposites.

I wish all the success in the attempt made by the editors and authors.

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PREFACE

Smart polymeric nanocomposites have attracted the attention of both scientists and researchers for the last few decades as they confer the materials with virtuous processability and sensitivity towards many external stimuli such as light, temperature, pH, electrolyte concentration, magnetic field, *etc*. In the dynamic landscape of materials science, the field of smart polymeric nanocomposites stands at the forefront of innovation, offering a spectrum of possibilities that outdo traditional material boundaries. These smart polymer nanocomposites find applicability in a wide range of applications such as sensors and actuators, drug delivery, self-healing structural material, smart textiles, smart electronics, aerospace applications, *etc*.

This book provides a broad updated survey and information on major categories of smart polymeric nanocomposites such as self-repairing materials, shape-memory materials, stimuli-responsive materials, *etc*. Synthetic routs and applications of all these materials are discussed. The essence of this book is centered on its dedication to encouraging inventive methods in tailoring polymeric nanocomposites to meet the characteristic requirements of cutting-edge applications. From the fields of biomedicine and wastewater treatment to the forefront of self-healing materials, food packaging, sensing technologies, and drug delivery systems, the book reveals a wide range of opportunities where intelligent polymeric materials take on a crucial role.

This book is a culmination of the collective expertise of scholars and experts from around the globe, exploring the fundamental principles that strengthen the design and utilization of smart polymeric nanocomposites. We are highly grateful to all contributing authors/ coauthors who contributed to this book and made our journey successful. We are also grateful to the Bentham Science Publication and its support team for helping us during the completion of this book.

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CHAPTER 1

Smart Polymer Nanocomposites: An Overview

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Abstract: Smart polymer nanocomposites have gained a wide range of applications in diverse fields of science and technology as they confer the material's great processability and sensitivity to a wide range of environmental stimuli, including magnetic fields, light, temperature, pH, and electrolyte concentration. Incorporating functional or pristine nanoparticles/nanofillers into polymer matrices significantly modifies the properties of smart polymers, including their shape memory ability, self-healing ability, pH sensitivity, thermal responsiveness, electro responsiveness, *etc*. These smart polymer nanocomposites find applicability in a wide range of applications such as sensors and actuators, drug delivery, self-healing structural material, smart textiles, smart electronics, aerospace applications, *etc*. This chapter seeks to address the importance, synthetic strategies, characterization techniques, and application of smart polymeric nanocomposites, summarizing an overview of the range of these materials.

Keywords: Actuators, Diels-Alder reaction, Graphene, *In-situ* polymerization, Melt-processing, Nanoparticles, Nnanocomposites, Nanotubes, Pi-Pi interaction, Solution-blending, Self-assembly, Smart polymers, Stimuli-responsive polymers.

INTRODUCTION

Smart materials, which are also called stimuli-responsive materials, have gained considerable interest among the scientific community in the last few decades owing to their unique properties of sensitivity towards many external stimuli such as light, temperature, pH, electrolyte concentration, magnetic field, *etc*. Polymers are the most extensively used materials for developing stimuli-responsive materials as their chemistry allows easy modification of the properties by integrating functional chemical moieties. On the other hand, there are many polymers such as Poly(N-isopropyl acrylamide), Poly(acrylic acid), Poly(ethylene glycol), Poly(2-hydroxyethyl methacrylate), Polyethyleneimine, *etc*., which themselves are responsive towards external stimulus [1 - 4]. These polymers can detect one or more stimuli as a signal, determine the extent of the change, and then directly respond to the stimulus by changing the conformation of their

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chains. These responses show up as alterations in the solubility, volume, shape, degree of association, and other properties [5, 6].

Polymer nanocomposites are materials that combine organic and inorganic components with at least one filler dimension less than 100 nm. Due to the increased interfacial area between the polymers and the nano-fillers, they exhibit better mechanical and physical characteristics than the host polymers [7 - 9]. Incorporation of pristine or functional nanoparticles into a polymer matrix can drastically enhance and modify the properties due to their high surface area, reinforcing effects, nucleating effects, and intrinsic functionalities. These nanomaterials can be derived from special polymers such as shape memory polymers, stimuli-responsive polymers, electro-responsive polymers, self-repairing polymers, self-cleaning polymers, sensingpolymers, *etc*. directly or by adding responsive nanoparticles. Resulting smart nanocomposites have a wide range of applications such as sensors and actuators [10], drug delivery [11], self-heling structural material [12], smart textiles [13], smart electronics [14], aerospace applications [15], *etc*.

In the last few decades, numerous efforts have been made for the integration of properties of nanoparticles and polymers for designing stimuli-responsive materials. Hence, a large number of organic and inorganic nanofillers have been studied that can amplify or generate stimuli-responsive behaviour. Based on their responsive behaviours, nanofillers can be classified into two categories [16 - 18]. The first category of nanofillers is called inert nanofillers, which have no stimuli-responsive behavior. These include cellulose, silica, and clay materials. The second type includes carbon nanotubes, graphene, metal oxide nanoparticles, *etc*., which show stimuli-responsive behaviour along with reinforcement. These can be termed active nanofillers. For the synthesis of smart polymer nanocomposites, the second type of nanofillers is primarily used due to their ability to respond to different stimuli. These nanofillers interact with polymer matrix mostly through weak intermolecular force of attraction. Due to the high surface area of nanofillers, which favors agglomeration, a good distribution of nanoparticles in the polymer matrix is difficult to achieve. Hence, fabrication techniques of polymer nanocomposites have a profound effect on the final composite. Different synthetic strategies are used for this purpose including, melt processing [19], solution blending [20], *in-situ* polymerization methods [21], *etc*., which are discussed in detail in this chapter.

The aim of this chapter is to give an overview of various smart polymer nanocomposites showcasing diverse responsive behaviors such as shape memory effect, self-healing characteristics, pH responsiveness, and electro responsiveness.

Moreover, this chapter will focus on different synthetic strategies, characterization techniques, and utilization of these materials in various applications.

SMART POLYMER NANOCOMPOSITES

Shape Memory Polymer Nanocomposites

Biocompatible progression in polymer properties has led to the development of materials capable of adapting their physicochemical properties and/or structural conformations in response to the surrounding environment, analogous to those found in natural organisms known as "smart polymers" or "stimuli-responsive polymers". Shape-changing polymers are a class of stimuli-responsive polymers that undergo molecular or visible-level shape alterations [22]. In contrast, shape memory polymers (SMPs), another type of polymer, retain their original shape after a brief applied force is removed. The dual-shape memory polymer effect is the change in the shape of SMPs from a permanent to a temporary state upon application of a transient applied force [23]. The three stages of strain that shape memory polymers (SMPs) go through are: (i) applying strain above the glass transition temperature; (ii) deforming to the desired shape close to room temperature; and (iii) removing the force such that the polymer returns to its initial shape above the glass transition temperature [24]. The active controllable temperature of SMPs can be sustained through changes in composition and degree of cross-linking.

Various reinforcements are incorporated into the SMPs to enhance the recoverable strain. According to studies, adding nanostructures to polymers greatly enhances their interface characteristics, while the strength and modulus are increased by the microstructure [25, 26]. Consequently, the utilization of nanofillers like nanoparticles, nanotubes, and nanowires in SMP nanocomposites is gaining importance. For instance, the addition of nano-sized SiO_2 to polylactide-based polyester enhances recovery temperature, flexibility, and mechanical strength, making it suitable for biomedical applications [27]. Likewise, the incorporation of carbon nanofibers into epoxy-based shape memory nanocomposites improves Young's modulus and product strength [28]. There have been several studies published in which SMP nanocomposites are fabricated on resins using a variety of methods, including resin transfer molding (RTM), vacuum-assisted resin transfer molding (VARTM), filament winding, and resin film infusion (RFM). In the following section, different nanofillers used for the reinforcement of SMPs will be discussed.

CHAPTER 2

Shape-Memory Polymer Nanocomposites and their Applications

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Abstract: Shape-memory polymer nanocomposites (SMPCs) are considered an advanced type of smart material, in which a specific stimulus influences the structure and thus memorizes their unique shape. SMPCs are widely used in high-performance water–vapor permeability materials, sensors and actuators, intelligent medical devices, morphing applications, and self-deployable structures in spacecraft. Modification of SMPCs overcomes the limitations of the pristine polymer and classical composites and can be counted among the main groups of smart materials. The performance of SMPCs under thermal, electric, optical, magnetic, and solvent stimuli can be improved further, potentially giving them a new range of uses. The most extensively researched SMPCs are those based on polyurethane, epoxy, polycaprolactam, polylactic acid, and polyvinyl alcohol; these also include those nanofillers based on carbon like CNTs, carbon black, graphene oxide, graphene nanoplatelets, graphene quantum dots, *etc*; metal oxides like Fe_3O_4 and TiO_2; cellulose like cellulose nanocrystals and nano-cellulose gel; and other nanomaterials such as nano-clay, TiN, AuNRs, organic nanoparticles, silica, sepiolite, silsesquioxane, and hydroxyapatite nanofillers.

Keywords: Advanced materials, Actuators, Fillers, High performance, Intelligent medical device, Nanoparticles, Nanocomposites, Sensors, Stimuli-responsive, Shape memory effect, Shape memory polymer, Smart materials, Self-deployable, Thermo-responsive.

INTRODUCTION

Shape-memory polymers (SMPs) are considered a kind of smart material with tunable size, shape, stiffness, and strain in response to different external stimuli (like heat, electric and magnetic field, water, or light) including physiologic ones such as pH, body temperature and ions concentration. The unique property of shape memory polymers is that they have the capability of storing and recovering

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a large deformation. Furthermore, SMPs are materials with the ability to sense external stimuli and react in a predictable and regulated way. They can be designed to memorize one or more temporary shapes in addition to a permanent shape [1, 2].

A polymer needs both the switch segment and the net point in order to display the shape memory property. The initial shape can be created by the crystalline phase, by cross-linking *via* a covalent connection, or by physical means and an interpenetrated polymer network that is determined by the net point of SMPs. The net point must remain constant through the formation of a stable polymer structure in order to maintain the material's shape even at high temperatures. The switch segment, in particular the flexible portions of the polymer matrix for reversible switching transition, can be a physical transition and can restore the temporary structure of SMPs. Moreover, a physical phenomenon like crystallization or glass transition of the polymers and chemical reactions like oxidation or reduction, Diels–Alder reaction, supramolecular association or disassociation, self-assembly of metal-ligand coordination, *etc*., influence the shape memory properties of the SMPs [3, 4]. Based on the development of molecular mechanisms, the comprehensive architecture of the SMPs is presented in Fig. (**1**) [5].

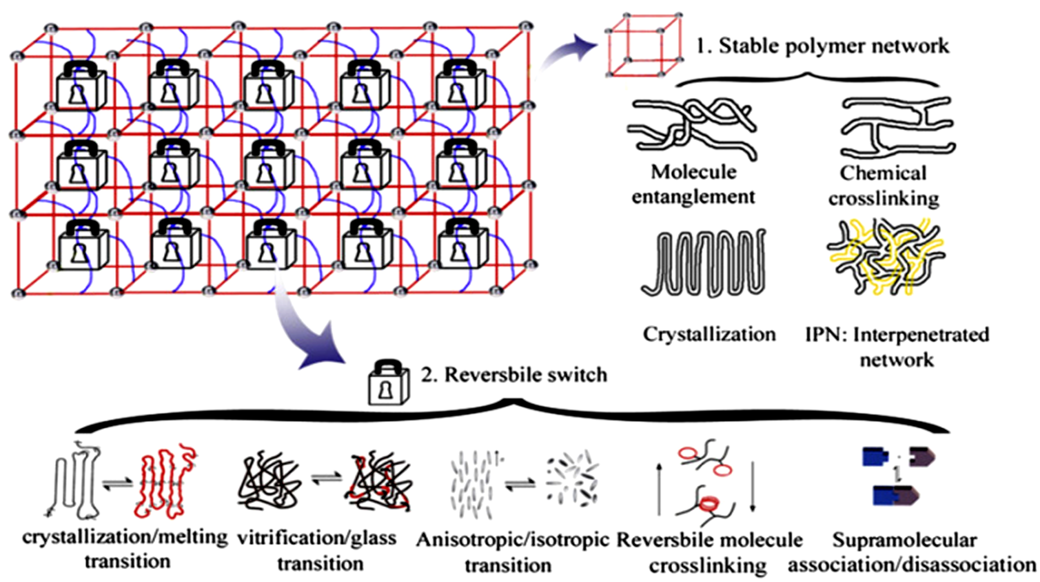

Fig. (1). Various molecular structures of SPMs [5].

SMPs are frequently used in high-performance water-vapor permeability materials, self-deployable structures in airplanes, morphing applications, textiles and clothing, sensors and actuators, and smart sophisticated medical equipment. However, the high-performance applications of SPMs are limited by low heat conductivity, low recovery stress, low tensile strength and stiffness, and inertness to electrical and light stimuli [3, 4]. Among all the SMPs, epoxy, polyurethane, polylactic acid, polycaprolactam, and polyvinyl alcohol-based SMPs are widely studied.

Due to nanofillers' strong polymer interaction and wide surface area, researchers have focused more on shape memory polymer nanocomposites (SMPCs) recently than on traditional composites. This way of preparation of SMPCs is an effort to address the shortcomings of SMPs [6]. Nanofillers such as metal oxide, nanotubes, nanofibers, nanospheres, nanorods, and so on can be incorporated into a polymer matrix to create a variety of SMPCs with specific properties. The decoration of SMPCs can enhance the performance characteristics of the SMPCs under electric, light, magnetic, thermal, and solvent stimuli, which may open up a new dimension for the SMPCs to be used in more advanced applications [5, 7].

Meticulous designing of SMPCs can improve the performance characteristics of the SPMs and increase the adaptability of stimuli-responsive properties. By adding a modest amount of cross-linking micro- or nano-sized fillers, the mechanical strength and recovery stress of SMPs can be significantly increased. Hence, as compared to the tidy SPMs, the designed SMPCs show noticeably better mechanical and shape memory qualities along with the new properties. Decoration of SMPCs can improve thermal stimuli-active and shape memory properties along with the reinforcement effect of the fillers. Typical examples of unique shape memory effects include stimuli-memory effect, two-way shape memory, multiple-shape memory, spatially controlled shape memory, and self-healing [7].

The fillers in SMPCs can improve the way SPMCs perform by activating the polymer matrix with different energy inputs and giving the composite system varied functions. But there are still certain obstacles to overcome. Due to the substantial or cyclic deformation that SMPCs endure, the primary problem in the manufacture of SMPCs is achieving homogeneous mixing of the fillers and SMP matrix. The mechanical characteristics of SMPCs are diminished by the modest intermolecular forces of attraction between the fillers and the matrix. Additionally, this may result in the Mullins effect of filled composite materials, which lowers the mechanical qualities of the polymer composites that are the outcome of the first extension. The overall performance characteristics of the SMPCs are influenced by the filler's geometries, modulus, volume contents, CHAPTER 3

Smart Polymeric Composites in Controlled Drug Delivery

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Abstract: A drug delivery system (DDS) is the mode by which a drug is incorporated inside the body to produce therapeutic effects at the desired pathological site. Common DDSs include capsules and tablets (through the oral route), intravenous injections, nasal and ocular drops, transdermal patches, ointments, gels, *etc*. The important criteria for an active pharmaceutical ingredient (API) to be effective are that it should be precise and specific for the site of interest, with minimal side effects. This is where the formulation of the drug becomes very important. Regulating the entry of the drug into the body, followed by excellent distribution is necessary to elucidate a quick reaction. Hence, substantial consideration has to be given to the absorption, distribution, metabolism, and excretion (ADME) of the drug. Polymers have been integrated with APIs mostly as inert drug carriers to facilitate ADME, with the pre-requisites that the incorporated polymers must be biocompatible, have the ability to improve the drug stability and release kinetics, and avoid accumulation in the bloodstream. To further improve their efficacy, smart polymers have also come into play. They react to environmental stimuli like temperature, light, magnetic and electric fields, *etc*. Polymeric magnetic nanocomposites have shown great promise due to their quick response to magnetic cues and remote-control capabilities. The functionalization of both natural and synthetic biodegradable polymers with superparamagnetic iron oxide nanoparticles (SPIONs) results in DDSs where drugs can be remotely released in a controlled manner.

Keywords: Chemically controlled, Core-shell structure, Diffusion-controlled, Drug delivery system, Magnetic hyperthermia, Magnetically-guided delivery, Multi stimuli-responsive, pH-responsive, Polymer-drug carriers, Smart polymers, Solvent-activated, Superparamagnetic iron oxide nanoparticles, Temperature responsive.

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INTRODUCTION

Overview of Drug Delivery Systems

A drug delivery system (DDS) is the method of administering a pharmaceutical formulation inside the body to realize a therapeutic effect. Once a novel drug is developed in the laboratory, the chances of it being solely administered are very rare. Instead, the drug (or more aptly called the active pharmaceutical ingredient (API)) is given as a part of a formulation, combined with inert pharmaceutical ingredients called excipients. The excipients serve very specialized functions and are a part of the drug delivery process. Thus, a DDS can also be described as a means for the drugs to be packaged in such a way that they are protected from physiological degradation and can be delivered at the target site safely at a controlled rate. DDSs can be categorized in two ways: (a) based on the mode of administration of the drug into the body and (b) the intended effect at the target site upon delivery. Both the mode of administration and the intended effect can be local or systemic [1]. Local administration of the DDS is mainly achieved by directly applying it to the target site or introducing it into the circulation system in such a way as to obtain a high concentration of the API at the desired location without exposing other body parts. Local administration routes mainly include applying topical ointments, lotions, creams, and gels transdermally, and also through nasal and eye drops, *etc*. Although advantageous, local administration in most cases is difficult due to accessibility issues arising out of anatomical barriers. In such cases, systemic administration is required where the DDS is applied at an off-target accessible location *via* oral tablets and capsules, intravenous injections, *etc*. The DDS then circulates through the body to reach the intended target site. Due to circulation, the possibility of non-intended organs getting exposed to the drugs increases, leading to higher chances of side effects and toxicity. Once delivered, the drug can produce either localized or systemic effects. Localized effect entails drug activity typically at the site of DDS administration (local effect by local administration). However, it is also possible to obtain local effects from systemic administration by causing the DDS circulating in the bloodstream to accumulate at and affect the target site. If the drug affects the entirety of the body during circulation, this is termed a systemic effect by systemic administration.

During the initial days of therapeutics, APIs were mainly created using small molecules (sizes < 900 Daltons) like steroids and antibiotics. They can effortlessly diffuse through various biological membranes but for that to happen, they must dissolve easily in the biological fluids, which is often not the case. As the therapeutic landscape evolved, other classes of APIs were introduced, including proteins and peptides, antibodies, nucleic acids, live cells, *etc* [2]. These drugs serve new therapeutic functions, but they are also riddled with some difficulties of their own, like problems with stability, delivery conditions, *etc*. In response to the hardships, DDSs have also advanced in order to solve these solubility and stability issues by modifying the local microenvironment and physicochemical properties, and also improve bioavailability, and broadening the activity of the API. An important component of the DDS that is instrumental in solving these issues is the excipient. Additionally, some excipients also act as drug carriers. As the name suggests, a drug carrier acts as a protective barrier for the transportation of pharmaceutical molecules to the target site, without causing any degradation or leakage during the entire process of administration, circulation, and delivery of the drug. An ideal drug carrier should have the ability to control and optimize the rate of drug delivery and increase its duration of activity so that the frequency of drug administration can be reduced.

Smart Polymers as Drug Carriers

Biocompatible polymers have been widely used in DDSs in various ways. They can either be non-biodegradable or biodegradable. In the case of non-biodegradable polymers, surgery is often required to excrete them out of the body once their purpose is completed. Hence, the use of biodegradable polymers is favored. Biodegradable polymers decompose into non-toxic and regular metabolic products through hydrolysis or enzyme-catalyzed reaction. They are classified into two categories: natural polymers, such as cellulose, chitosan, polypeptides and proteins, and synthetic polymers including poly (glycolic-co-lactic) acid (PLGA), poly (lactic acid) (PLA), poly (glycolic acid) (PGA), poly (caprolactone) (PCL), poly (ethylene glycol) (PEG), poly (vinyl pyrrolidone) (PVP), poly (methyl methacrylate) (PMMA), aliphatic polycarbonates and polyphosphazene. Polymers incorporated with drugs can modify the circulation characteristics of the drugs, and degrade at the target site to improve drug release and avoid carrier build-up. Additionally, polymers themselves can sometimes be bioactive and offer therapeutic advantages. Synthetic polymers are preferred over natural ones since it is easy to control the polymer's three-dimensional structure and chemical composition during synthesis. In that way, polymers can be tailor-made to fulfill desired requirements. For instance, polymers can be synthesized with their functional group orientation in a particular direction so that they interact strongly with the API. Synthetic polymers, however, are not always degradable. In such cases, they must be eliminated from the body through renal excretion. So, consideration has to be given to maintaining their molecular weight under the renal excretion threshold.

Polymers can be incorporated with the APIs in a variety of ways: polymers can form covalent bonds with the drug to form a polymer-drug conjugate, or the drug can disperse in the polymer network to form a matrix system, or the drug core can

CHAPTER 4

Environmental Applications of Smart Polymer Composites

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Abstract: The growing population and depletion of fossil fuels not only hamper industrial system, but also affect everyday life. Rapid urbanization, change in human lifestyle and continuous production of toxic chemicals are directly increasing the amount of chemical waste, air/water polluting substances, which are causing hazardous affect to human health for which no adequate solution is available. Thus, developing new technologies has been a priority for researchers to resolve such issues. Currently available conventional technologies need further development or replacement with newer technologies. Substantial research efforts put forward different methods among which the use of smart polymer materials to reduce environmental problems is observed to be one of the best. Intelligent polymer materials which are strategically designed to conquer multiple properties in diverse conditions such as pH, temperature, stress or electric field made their presence necessary. The response rate of these materials against different environmental stimuli can be regulated by simple functionalization, leading to significant industrial and domestic applications. In this chapter, the readers will get an overview of the using smart materials for environmental issues, and possibilities of exploring further contributions for solving environmental challenges.

Keywords: Antifouling, Catalysis, Composite material, Environment, Sensing, Self-healing, Smart polymer, Water harvest.

INTRODUCTION

The impact of growing modern civilization and rapid changes in human lifestyle concerns the government and the scientific community due to its harsh involvement towards environmental problems [1]. At various times, incidents like the Kalamazoo river oil spill and Huangdao oil pipeline explosion have shown disastrous moments when smart materials played a crucial role in overcoming that situation. Governments from different countries worldwide have already issued alerts regarding the impact and resolutions to solve these environmental problems.

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Therefore, advancement of materials/methods for detecting, preventing, and remedying environmental damage is essential to socio-ecological health.

The materials that sense and respond to changes in their surroundings in a very precise and reproducible manner are called Smart Materials [2, 3]. They are considered environmentally significant due to their several environment-driven applications including catalysis chemistry, sensor chemistry, and chemistry relevant to decontamination methodology that is relevant to environmental problems [4 - 9]. The application in catalysis chemistry is of utmost importance because of the ecological concerns of producing chemical and hazardous wastes. Sensor chemistry is essential due to misusing toxic materials for specific purposes. Again, negligence, carelessness, and ignorance in handling carcinogenic and toxic chemicals result in polluting research/industrial facilities, water reservoirs, and the atmosphere. Researchers worldwide are engaged in identifying materials that can act swiftly, and could be implemented to reduce various types of waste produced in chemical processes, detect the presence and concentration of contaminants, and decontaminate fouled environments.

In **a** recent study, poly(vinyl chloride) (PVC) membrane-based sensors were utilized to investigate the presence of antipsychotic agents [10]. In this study, El-Ragehy *et al*. constructed modified electrodes using tetraphenylborate or tetrakis (4-chlorophenyl) borate for selective detection of fluphenazine and nortriptyline hydrochlorides *via* ion-pair formation. Another poly(vinyl) chloride (PVC) membrane electrode was prepared by Liu *et al*. [11]. This electrode comprised of pethidine–phosphotung is used for the determination of pethidine hydrochloride drug in injections and tablets. Besides polymer materials, nanocomposites prepared by incorporating nanomaterials into polymeric materials are also known for their novel applications. In this chapter, we have discussed briefly about the different environmental applications of smart materials. We hope that the readers will get an overview of the recent developments and future endeavours of smart materials for solving ecological concerns.

ENVIRONMENTAL APPLICATIONS OF SMART POLYMER MATERIALS

The amalgamation of environmental science with material science unlocked the door to overcome future challenges. The use of smart polymer materials for environmental benefits has been the focus of research and development for quite some time. The use of smart polymer materials involving catalysis chemistry, sensing chemistry, and decontamination methodology is especially relevant in addressing different environmental challenges [4, 5]. Furthermore, smart polymer materials are also utilized for self-healing, antifouling, water harvesting, *etc*., some of which are discussed in this section.

Catalysis

Catalysis plays a significant role in synthetic laboratories and industrial processes. Catalysis is categorized into two types: heterogeneous (substrate and catalyst state are different) and homogenous catalysis (substrate and catalyst state are same). In the race to develop sensitive, and cost-effective, and reliable catalysts, smart polymer-based materials withdraw much attention due to their enhanced stability, uniform dispersion behavior and effortless functionalization properties [4]. For example, Xiao *et al*. studied the use of temperature/pH-responsive gold nanoparticle-incorporated hybrid nanogels for catalytic reduction of 4-nitrophenol. The catalytic efficiency was significantly affected by tuning the temperature, kinetics, and thermodynamics of the chemical reaction [12]. In another example, a microgel system composed of Nisopropylmethacrylamide and methacrylic acid (MAa) encapsulated with bimetallic (Ag/Ni) nanoparticles was used to catalyse the degradation of different azo dyes (Fig. **1a**) [13]. Temperature-responsive smart catalytic systems are also well known for their high catalytic performance at specific temperatures. A graphene-supported nanocatalyst $Au_{core}Pt_{shell}$ bimetallic (G-Au@Pt) was used for the catalytic reduction of 4-nitrophenol. In the presence of $NaBH_4$ the catalytic activity, kapp = 9.44×10^{-3} s^{-1} was higher than the previously reported catalysts (Fig. **1b**) [14]. Also, a temperature-dependent catalytic study revealed that the catalyst performance was linearly increased in the temperature range of 15-30 ºC. However, the catalytic performance dramatically drops on increasing the temperature, calling it "switched off" (<35 ºC).

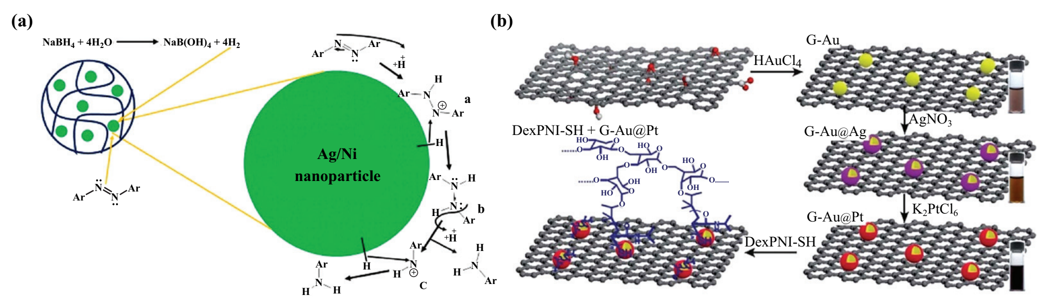

Fig. (1). (**a**) Plausible mechanism of catalytic degradation reaction of MOr using Ag/Ni-P(NMA-MAa)and $NaBH_4$ [13]. and (**b**) Synthesis of modified graphene sheets with $Au_{core}Pt_{shell}$ nanoparticles and attachment of the dextran-based smart polymer with thiol-ends [14].

CHAPTER 5

Application of Smart Polymeric Composites/Nanocomposites in Drug Delivery

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Abstract: Over the course of several decades, an interest in stimuli-responsive polymers has grown significantly, leading to extensive research in the development of environmentally responsive macromolecules, which can be exhibited as smart novel polymers. They have emerged as a promising platform for the advancement of drug delivery systems, enhancing the efficiency of therapeutic interventions, decreasing dosage frequency, minimizing possible adverse reactions and enhancing patient receptivity. In particular, these materials are responsive to very subtle changes in their environment. The environmental triggers for such transformations can include temperature change, shift in pH, variations of ionic strength, alterations in magnetic or electric fields, exposure to light, *etc*. They are said to be stimuli-responsive or intelligent polymers. Smart polymeric materials provide a versatile platform for drug delivery, enabling the controlled release of drugs in a stable and bioactive form. Additionally, the incorporation of nanocomposites contributes to increased drug bioavailability, enables targeted delivery through surface modification, and facilitates sustained release profiles. This chapter mainly provides detailed progress of smart polymeric composites or nanocomposites in the advancement of drug delivery applications. It also thoroughly covers several characteristics of these polymers categorized into different groups based on the stimulus type. The chapter aims to shed light on the evolving landscape of stimuli-responsive polymers and their pivotal role in shaping the future of drug delivery systems.

Keywords: Drug delivery, Intelligent polymers, Nanocomposites, Stimuli-responsive, Smart polymers.

INTRODUCTION

The polymers in today's world play a crucial role, and it is difficult to imagine life without them. They are a group of advanced materials with uses in various areas

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of science, technology and industry – from fundamental applications to biopolymers or therapeutic purposes. Polymers are available naturally and are also man-made. The study of man-made polymeric materials became very prominent during the mid-19th century. Since then, the polymer industry today has seen rapid growth and is now greater than the combined industries of steel, copper, aluminum, *etc* [1].

A polymer is a large molecule (macromolecule) made up of long chain repeating units of small molecules. These small molecules are usually connected by covalent chemical bonds and these repeating units are called monomers [2]. The awareness of various kinds of polymers and their applications among people emerged only after World War II [1]. Depending on the type of molecules bonded, they have unique properties and applications. Polymers in today's world have found applications ranging from transportation to medicine and electronics, clothes and cookware to housing, *i.e*. the impact of polymer in the present way of life is immeasurable [3]. Because of advantages such as ease of processing, better strength-to-weight ratios, durability against chemical and physical degradation, and cost-effectiveness, polymers have replaced a number of conventional materials in various applications [4]. As such, polymers serve as one of the primary contributors to the industry as well as the economy. This is evident from the fact that Europe had a production volume of 57.9 million tons of polymers in 2019, which accounts for 16% of the world's total production with Asia being the leading producer [5].

The inspiration and intelligence of nature in recent times have led to the development of the smartness of material. The polymers that integrate the combined functions of sensing, actuation, and control can be defined as "smart polymers". Amidst the era of the 4th industrial revolution, researchers have drawn a huge interest in smart polymers [6]. These polymers respond to mild alterations in chemical as well as physical states with sharp and relatively large phase or property changes. The responses to external stimuli result in macro-scale changes like contraction, expansion, twisting, elongation, or rolling of the materials [7]. As such, they are also regarded as "environmentally sensitive" polymers. These polymers have the ability to take many forms, and can be physically or chemically modified with other bioactive molecules as well. Over the past 20-25 years, smart polymers have found extensive uses in the biomedical field [8]. Smart polymers in the medical field are used for the treatment of several diseases and also in advanced medical sectors. They respond to temperature, pH, enzymes, and concentration of analytes, light, or magnetic field. Smart polymers have also found applications in bioseparation, data storage devices, and rechargeable batteries to name a few. Thus, the emerging research in the field of smart polymer has not only opened avenues for their applications in the medical field but also in the food sector, architecture, data, and energy storage [9]. This chapter mainly focuses on the significant advancements made by smart polymeric nanocomposites for drug delivery applications, as it undergoes a thorough exploration, covering diverse aspects of these stimuli-responsive materials. It also offers comprehensive insights into their evolving role and impact on drug delivery systems, by highlighting the crucial contributions of these smart materials in drug delivery technologies.

POLYMER NANOCOMPOSITES

Due to the various properties of polymers and their composites like lightweight, low cost, excellent corrosion resistance, easy processing, *etc*., polymer composites have widespread applications in all aspects of daily life and industry [10]. Polymer composites are a combination of polymers and a material in which the final product has superior properties compared to each constituent material. A polymer composite usually consists of a continuous phase (matrix) and a discontinuous phase (reinforcement) [11]. Polymer nanocomposites on the other hand are polymer composites with fillers having any one dimension less than 100 nm. They usually have very minimal amounts of well-dispersed nanofillers compared to polymer composites [12].

SMART POLYMER NANOCOMPOSITES

Smart polymer composites and nanocomposites are an advanced class of materials, which respond to environmental stimuli like light, enzyme, pH, or electric field, resulting in better performance and increased efficiency. Table **1** shows some examples of stimuli-responsive polymers and their significant results in the biomedical sector [13]. Before the application of smart polymer composites and nanocomposites, there is a need for extensive research on the capabilities of smart polymer composites/nanocomposites to revolutionize several industries [14]. The ability of smart polymer composites and nanocomposites to respond to environmental stimuli provides various advantages like greater performance, increased efficiency, and self-healing capacity. Some widely used smart polymer composites and nanocomposites are shape-memory, self-healing, and conductive polymers. The shape memory polymers have the ability to revert back to their original character as the deforming force is withdrawn. Conductive polymers can conduct electricity and are used in applications such as actuators or sensors. Self-healing polymers are those, which can repair damage like cracks or fractures caused by external factors, thereby extending their lifespan [15].

CHAPTER 6

Smart Polymeric Nanocomposites for Water Purification

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Abstract: The process of water treatment is aimed at reducing or removing contaminants either chemical or biological in water for its utilization in safe drinking or any other essential purposes. Polymer nanocomposites that show responsiveness to external stimulants play a vital role in this process and purify water by actuating stimuli and by reviving the polymers successively. In response to environmental changes, these polymer nanocomposites can firmly undergo physical as well as chemical characteristic changes like shrinkage, and expansion, or show alterations in optical and electrical features on the basis of the induced stimulants. So far, numerous polymeric compounds that are sensitive to light, heat, CO_2, *etc*. including prefabricated nano polymers, nanoparticles, *etc*. have been employed in water treatment. This book chapter emphasizes the efficacy of different smart polymeric nanocomposites in removing harmful pollutants including toxic ions, water-soluble ions, and harmful organic colorants in the water treatment process.

Keywords: Dye, Heavy metal ions, Stimuli-responsive polymer, Water purification.

INTRODUCTION

In recent times, more emphasis has been given to research, to develop smart polymers that are sensitive to both internal and external stimuli [1 - 7]. These responsive polymers are counterfeit and produced with customised qualities so that they can exhibit preferred responsiveness to specified impulses [8]. In this book chapter, we have focused on the application of various responsive materials in water treatment, both domestic and wastewater. With the emergence of human progress, remarkable improvements in industry, and advancement in the agriculture sector, unplanned human activities are slowly causing water contamination, resulting in scarcity of fresh water. The toxic chemicals including

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heavy metal ions, organic dyes, and other pollutants from industry significantly damage water bodies. The need for a permanent solution to overcome this serious issue has led to the development of many outstanding technologies that involve liquid-liquid partitioning, ultra or microfiltration, use of zeolites, resins, and other adsorbents, use of chemicals for easy sedimentation, electrochemical technique, *etc* [9 - 11]. Among them, adsorption and membrane filtration attract researchers because of their highly effectual and specific nature in pollutant exclusion, the need for minimum energy, maximum outturn, easy handling, and trouble-free expansion [11, 12]. Past years have endorsed the tremendous application of smart substances mainly nanopolymers in wastewater refinement.

These polymeric actuators are seen to be highly effective in the purification of wastewater owing to their magnificent characteristics under divergent circumstances such as heat, radiation, pH, CO_2, magnetic field, and electric field. The introduction of these materials gives a new dimension to the water treatment process, as the purification process can be governed just by changing the nature of the applied stimuli. Also, these smart polymers are self-cleanable and do not have any fouling characteristics, which leads to an increase in their functional ability and are thus cost-effective. Herein, we discuss the advancement of such delicate materials, their fabrication, features, as well as application in aquatic purification until now.

Classifications of Smart Actuators Used in Water Treatment

Stimuli-responsive materials employed in aquatic remediation are either alert to particular impulses like heat, pH, UV-visible light, energy fields, electrolytes, and ionic compounds, *etc*., or numerous impetuses, working simultaneously, otherwise in turn. This section will discuss various responsive materials that have been applied till now for the cleansing of water.

pH-sensitive Polymers

Various scientific methods, such as physical adsorption [13], oxidation [14], bioremediation [15], and stimuli-responsive materials [16], *etc*., have been employed over the past decades to remove toxic materials like heavy metals, inorganic impurities, harmful dyes, *etc*., from groundwater as well as surface water. Among the various stimuli-responsive materials, the pH-responsive materials are widely studied for water treatment. With the variation of the pH, these polymers undergo modification in their exterior action, solubility, and the sequence of the components. Basically, in an acidic medium, these responsive materials act as cationic polymers, and in the basic medium, they behave as anionic polymers, indicating their different self-assembly nature, which depends on their structures. The ionic attraction between the counter ions and dyes that are absorbed by these polymers, which alters the pH of the state, is the key factor of their high removal efficiency for these harmful chemicals.

pH-Sensitive Cationic Smart Materials

Efficient removal of NO_3^- from the water was studied by Zahra Abousalman-Rezvani and co-workers [17], who synthesized grafting light, temperature, and CO_2-responsive copolymers from cellulose nanocrystals (CNC)-grafted and free block copolymers of 2-dimethylaminoethyl methacrylate (DMAEMA) and coumarin by atom transfer radical polymerization. The structural and thermal characterizations were observed by Fourier-transform infrared spectroscopy, proton nuclear magnetic resonance, and thermogravimetric analysis. With the increase in pH, the LCST of the polymer decreased. Bubbling of CO_2 through an aqueous solution results in protonation of the amine group of the polymer, thus adsorption of the ions increases, causing LCST at higher temperatures. Deprotonation of the polymer was observed on inserting N_2 gas. Yan Qin *et al*. studied [18] the removal of methylene blue (a-MB), a negatively charged acid dye, from water with high efficiency using pH-responsive ammonium-functionalized hollow polymer particles (HPP-NH_3^+) with a high density of ammonium groups in the shell. Remarkable pH-dependent equilibrium adsorption was shown by HPP-NH^{3+} polymers that increased from 59 mg/g to 449 mg/g with a decrease in the pH from 9 to 2. The reason behind the pH responsiveness of the polymers is the presence of the dual functional group (ammonium and carboxyl groups). HPP-NH^{3+} exhibited low adsorption capacity for b-MB while for a-MB the case was reversed, indicating its selective nature for acidic dyes. The adsorption equilibrium was attained at 15 mins with an adsorption capacity of 194 mg/g, having an initial a-Mb concentration of 200mg/L. The polymers can be easily recovered at basic solution pH = 10 and can be reused after 5 cycles with 98% dye removal efficiency.

Nowadays, biopolymer-based adsorbents draw more attention in the purification of aquatic systems as they are inexpensive and environmentally safe [19]. Among them, pH-responsive biopolymers are investigated by most researchers for the removal of harmful dyes from water. Bincheng Xu and co-workers reported a novel chitosan-based magnetic adsorbent [poly(2-(dimethylamino)ethyl methacrylate) grafted magnetic chitosan microspheres, GMCMs] and their efficient adsorption of dyes that are negatively charged, such as Responsive Blue 19 and Acid Green 25. The remarkable adsorption capacity of GMCMs was found to be at pH 2 and at a contact time of 90 min due to complete protonation of the grafted PDMAEMA chains, and they became positively charged electrolytes. Along with electrostatic attraction, the attractive force *i.e*., hydrogen bond, amid the polymers and the colorants also increased its adsorption capacity. The

CHAPTER 7

Stimuli-Responsive Polymeric Nanocomposites and their Applications

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Abstract: As technology globally embraces "smart" features, the polymer industry has acquired some advanced creations, including the development of polymer nanocomposites (PNCs) characterized by smart or intelligent properties. These smart features originate from the ability of these materials to respond to external cues such as pH, temperature, stress, light, electricity, magnetism, chemical environments, *etc*., facilitating their end use as flexible electronic or motion sensors, shape recognizable polymers, and self-healable materials. The sensitivity of the materials can be hierarchical within the polymer matrix or can be adventitiously introduced by the nanomaterials resulting in a combined system that reflects some additional characteristics. This chapter aims to highlight the various aspects of stimuli-responsive smart PNCs; including a brief overview of the fabrication methods of PNCs using different polymer matrices such as polyurethanes, polyesters, epoxy resins, hydrogels, *etc*., and various advanced metal- or carbon-based nanomaterials. The authentication methods as well as various sophisticated attributes of these materials are highlighted in the chapter. In turn, the numerous applications of these PNCs in real-world scenarios are also discussed briefly. Thus, the chapter represents an attempt to illustrate the fabrication methods of PNCs and their frontline applications that help in understanding their future possibilities in the area of material sciences.

Keywords: Drug delivery, Electro responsive, Magneto responsive, Mechanical, Nanostructure, Optical, pH-responsive, Photo-responsive, Polymer nanocomposites, Sensors, Solubility, Thermal stability, Thermo-responsive.

INTRODUCTION

Nature offers a remarkable array of examples showcasing stimuli-responsive behaviours in living organisms. Sunflowers exhibit heliotropism where the flower heads track the movement of the sun, chameleons change color in accordance to their environment, *Mimosa pudica* commonly known as the "touch-me-not" plant

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exhibits rapid leaf movement in response to touch, *Codariocalyx motorius* exhibits leaflet movement in response to various stimuli (light, touch, or sound); and many more. These examples highlight the remarkable adaptability and responsiveness of living organisms to their surroundings [1, 2]. Scientists draw inspiration from such natural phenomena to develop stimuli-responsive materials and technologies with applications ranging from robotics and biomimetics to smart materials and environmental sensing. Mimicking these natural mechanisms in synthetic materials not only led to significant advancements in the field of materials science but also opened up new avenues for innovation in various fields [3].

Polymers play a crucial role in material science and their characteristics showed great potential to yield smart materials. Thus, polymers have become one of the main research directions among stimuli-responsive materials since they have the ability to give remarkable responses to small changes. Living organisms showing stimuli-responsive behaviour are made up of natural polymers such as proteins, polysaccharides, and nucleic acids, and to mimic these natural mechanisms of stimuli responsiveness, synthetic polymers have been developed with similar functionalities [4]. Synthetic polymers can be classified into different categories based on their chemical properties, structure, and behaviour; such as thermoplastics, thermosetting polymers, elastomers, biodegradable polymers, smart polymers, *etc*. Continued research and innovation in polymer science and engineering hold the potential to further expand the range of applications of synthetic polymers as stimuli responsive polymers (SRPs). SRPs are also called stimuli-sensitive, smart, environmentally sensitive, and adaptive polymers [5, 6]. These polymers typically consist of repeating units with functional groups that can undergo reversible changes in conformation ensuing change in their physical or chemical properties in response to external stimuli. The most common stimuli that trigger responses in these polymers include temperature [7], light [8], pH [9], electric field [10], magnetic field [11], and oxidation-reduction [12, 13]. Different responses can be observed by the SRPs depending on the specific properties being altered and the above-mentioned stimuli triggering these changes (Fig. **1**). SRPs change the overall polymer chain dimensions or size by inducing the expansion or contraction of polymer chains. For example, temperature-responsive polymers may undergo coil-to-globule transitions, where the polymer chains either collapse or swell in response to changes in temperature [14]. Some SRPs exhibit changes in their secondary structure in response to stimuli such as temperature, pH, or solvent composition. For instance, temperature-sensitive polymers may undergo transitions between ordered and disordered states, affecting their mechanical properties or interactions with other molecules [15]. Further, SRPs can undergo changes in solubility or dispersibility in solution upon exposure to specific stimuli, which may involve transitions between soluble and insoluble states, leading to phase separation or precipitation of the polymer. SRPs may also undergo changes in the degree of intermolecular association or aggregation in response to stimuli. For example, pH-sensitive polymers may form aggregates or micelles in response to changes in pH, affecting their behavior in solution. The reasons for the above-mentioned responses often involve the formation or disruption of secondary forces such as hydrogen bonding, hydrophobic interactions, or electrostatic interactions; simple acid-base reactions of moieties branched to the polymer backbone, conformational changes, crosslinking/degradation reactions, or phase transitions leading to the formation of new structures or morphologies [16, 17].

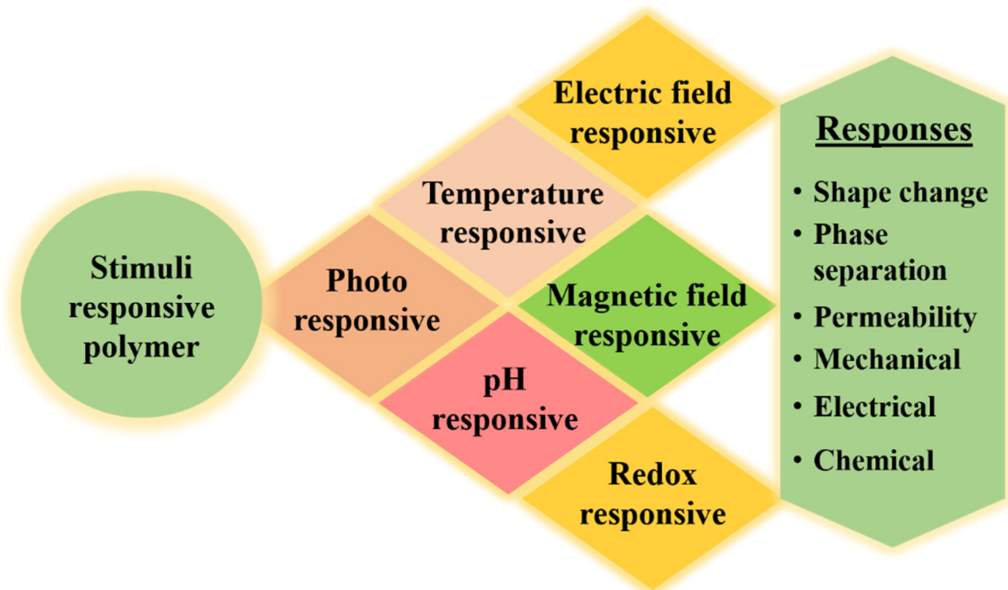

Fig. (1). Potential stimuli and responses of SRPs.

The stimuli sensitivity can be introduced into a polymer or stimuli responsiveness of SRPs can be enhanced by modifying the pristine polymer backbone chemically via the incorporation of multiple responsive units such as small polymer fragments or functional groups with high stimuli sensitivity. Further, nanomaterials can also be incorporated into polymer matrix leading to the formation of polymer nanocomposites (PNCs) either to add stimuli sensitivity into the polymer or to enhance stimuli responsiveness of the SRPs along with other properties making them suitable for practical use [18]. The high specific surface area, reinforcing effects and inherent functionalities of the nanomaterials help in introducing and enhancing stimuli responsiveness in the pristine polymers and SRPs, respectively along with enhancement of the overall performance. The

CHAPTER 8

Self-Healing Polymer Nanocomposites and their Applications

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Abstract: Self-healing materials can autonomously repair damage or cracks through dynamic mechanisms. These are designed in such a way that they are associated with a self-repairing mechanism supported by grafted functional groups on the backbone of the polymer. Among the self-healable polymer materials, polymer nanocomposites have drawn much attention in recent years in the growing field of materials science. This chapter is based on the methods of synthesis of self-healing polymer nanocomposites and provides a comprehensive discussion of the applications of self-healing polymer nanocomposites. Owing to their long-term stability and self-healing ability, self-healing polymer nanocomposites are considered one of the best composite materials among the emerging smart materials. Polymer nanocomposites are widely used in electronics, textiles, protective paint and coatings, biomedical, structural materials, *etc*. This chapter is based on the methods of synthesis of self-healing polymer nanocomposites and provides a comprehensive discussion of the applications of self-healing polymer nanocomposites.

Keywords: Biomedical, Construction materials, Electronics, Polymer nanocomposites, Self-healing polymer, Textile.

INTRODUCTION

Among the smart polymeric nanomaterials, self-healing polymer nanocomposites have garnered considerable attention in both research and day-to-day applications [1, 2]. A self-healing material composed of a polymer matrix reinforced with nanoparticles, designed to possess self-healing properties at the nanoscale level. Inspired by nature's resilience and biological systems, self-healing material can autonomously repair damage or cracks through dynamic mechanisms [3].

Self-healing polymer nanocomposites have profound applications in wide-ranging fields such as electronics, protective coatings, biomedical, membranes, structural

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materials, *etc* [4 - 6]. Self-healing polymers are designed in such a way that these are associated with a self-repairing mechanism supported by grafted functional groups on the backbone of the polymer. Such a functionalized polymer forms reversible bonds with different stimuli and possesses a significant advantage in the sense of physical or chemical response toward self-healing [7]. Thus, self-healable polymer nanocomposites have received much attention in recent years in the growing field of materials science [8].

The important characteristics of chemically functionalized polymer nanocomposites lie in their capacity to generate responsive interactions to various stimuli, including light, current, heat, light, *etc*. facilitating self-healing processes [9]. Moreover, self-healing polymer nanocomposites are cost-effective promising alternatives to prepare long-lasting polymer materials and great stands to triumph over environmental issues caused by plastics [10]. Therefore, these materials have considered more durable and eco-friendly materials for R & D and industries. Compared to other conventional materials, self-healing nanomaterials have an extreme ability to self-heal or self-repair when it is subjected to stress-causing strain. The research on self-healing material focused on the discipline of polymer and polymer nanocomposites, which are used in industrial applications and day-to-day life [11, 12].

On the basis of the synthesis and chemistry of self-healing polymer materials, they are classified as (i) autonomic and (ii) non-autonomic self-healing polymers [13, 14]. Autonomic self-healing polymer materials possess the ability to attach chemicals automatically, releaserepair, or heal the response to damage or rupture without any external intervention. The non-autonomic self-healing polymer materials on the other hand, repair or heal the response to damage or rupture in the presence of external stimuli. Resin-based polymers, incorporated as a healing agent in composite materials used in aerospace are non-autonomic materials. Moreover, self-healing polymer materials are also categorized as extrinsic and intrinsic self-healing materials based on their chemistry and applications. Intrinsic polymer materials can repair the damage without external intervention and extrinsic materials can be triggered by the incorporation of external additives to repair the damage or rupture [15].

Self-healing polymer nanocomposites have superior properties like strength, low cost, thermal stability, stiffness, lightweight, *etc*., which make them potential components in a number of emerging applications [16]. The successful application of self-healing concepts in the manufacture of polymer nanocomposite materials shows a new direction towards sustainability and development in a wide range of fields such as electronics, protective coatings, biomedical, membranes, long-lasting structural materials, *etc* [17].

This chapter discusses the methods of synthesis of self-healing polymer nanocomposites and provides a comprehensive discussion of the applications of self-healing polymer nanocomposites.

DESIGNING OF SELF-HEALING POLYMER NANOCOMPOSITES

A polymeric material shows self-healing properties which are associated with the ability to sense the force causing its damage, and begin the healing process autonomously (without further external stimulus) at the damaged site. A self-healing polymer heals the damage physically or by a combination of physical and chemical processes.

The key steps of all physical processes to design self-healing polymers are the inter-diffusion and entanglement of polymeric chains. Both properties are based on intermolecular forces, which are closely associated with the chemical characteristics of the polymer and the length of the polymeric molecules. Molecular inter-diffusion is induced by Brownian or chain segment motion, entropy-driven movement of particles or molecules, *etc*. in the presence or absence of an external stimulus. Several self-healing mechanisms like welding, patching, swelling, or *via* incorporation of nanoparticles can be assigned to the category proceeding through molecular inter-diffusion.

Among these, the use of nanoparticles to generate self-healing polymeric materials represents an interesting approach to healing different mechanical damages, *viz*. crack (internal damage) or scratch (external damage). The working mechanism of the self-healing polymer nanocomposite materials does not involve breaking and reformation of the polymer backbones like the other physical processes. Instead, it is based on the movement of highly mobile nanoparticles to the damaged vicinity that causes restoring the mechanical strength and thereby healing the damage [18]. In fact, nanoparticles act as dispersants to fill up the damaged part of a polymer causing healing of the material [19]. A small fraction of the dispersed nanoparticles absorb electromagnetic radiation and convert the energy into heat. It results in the formation of new bonds at the damaged site executing self-healing of the material [20 - 23].

On the other hand, the chemical process of designing self-healing polymeric materials is based on covalent or supramolecular type network formation. In covalent network formation, chemical bonds are generated between functional groups resulting in the formation of a permanent or reversible network. On the contrary, supramolecular networks are generally reversible associations of polymers connected through supramolecular interactions [18].

CHAPTER 9

Smart Biopolymers and their Applications

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Abstract: Over the last few decades, polymers have gained lots of importance in everyday life due to their unique properties and wide range of applications. In recent years, biopolymers have received attention due to their features like biodegradability, biocompatibility, renewability, and inexpensiveness. They are also used as smart and active materials in medicine, the food industry, *etc*., as they exhibit responsiveness to factors like temperature, pH, humidity, light, electrical and magnetic fields, *etc*. Therefore, this chapter emphasizes an in-depth analysis of various techniques and methodologies employed in the preparation of smart biopolymers, their characterization, applications, and the challenges associated with their development and utilization.

Keywords: Active-biopolymer, Bioengineering, Drug delivery, Edible coating, Functional food, Plastic pollutions, Protein nanoparticles, Renewable materials, Sustainable materials, Smart-biopolymers, Tissue engineering.

INTRODUCTION

Biopolymers, or polymeric biomolecules, are mostly biologically degradable polymers and are produced using living organisms, especially plants. Biopolymers are usually renewable and sustainable materials as they are prepared from living organisms. Their monomeric units are covalently bonded together, which leads to larger molecular structures [1 - 3]. Plants, microbes, and other naturally available materials are major sources of biopolymers. Different renewable materials are used to produce varieties of biopolymers that possess various physical, chemical, and biochemical properties [3 - 5]. In our nature, we have varieties of naturally available biopolymers such as lignin, starch, cellulose, hemicelluloses, *etc*. Biopolymers that are developed from natural resources have been manufactured in large quantities [6]. The biodegradable nature of biopolymers is due to the presence of some specific enzymes as well as microorganisms such as bacteria, algae, and fungi that possess distinct degradable characteristics [3]. The biopo-

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lymers have oxygen and nitrogen atoms in their polymeric chain, which make their biodegradability very simple [7, 8]. During degradation, these biopolymers are converted into biomass, water, carbon dioxide, and other natural components. The biopolymers are responsible for holding cells together in tissues, providing signals to the cells [9 - 11]. They are also responsible for providing elasticity and hydration to the skin and lubricating gastrointestinal tracts as well as joints of our body. Due to their additional benefits and functionality, the biopolymer possesses anadvantage over traditional polymers. There are many applications of biopolymers. These biopolymers can be utilized for reducing the simulation of chronic inflammation, and other toxic effects in the human body [12]. Biopolymers, being biodegradable, also have a variety of applications in the field of bioengineering like tissue engineering, drug delivery, and wound dressing. They can also be used in food packaging material, municipal solid waste disposal that reduces plastic pollution, and emission of carbon dioxide, resulting in a sustainable environment [13 - 15]. These biopolymers are often termed smart or adaptable materials, as their structure can be manipulated by using enzymes at different stages of their lifecycle. Moreover, these smart biopolymers are sensitive to small changes in the environment in terms of temperature, pH, solvents, *etc*., and may undergo fast and reversible changes in their structure and hence also in their physical properties [16 - 19].

Biopolymers: A State of Art

Based on origin, biopolymers are classified into two major categories. These are natural biopolymers, and synthetic biopolymers. These biopolymers are mainly found in living organisms like plants, animals, microbes, *etc*.

Natural Biopolymers

Biopolymers that are extracted from living organisms are known as natural biopolymers; for example, polysaccharides and proteins. Polysaccharides are long-chain carbohydrate molecules, or monosaccharides connected through glycosidic linkage. The major functions of polysaccharides in the human body are energy storage and structural support. They are abundant natural biopolymers found in animals, plants, and other microorganisms. Starch, cellulose, chitosan, chitin, *etc*. belong to naturally occurring important polysaccharides. Similarly, proteins also belong to an important class of biopolymers that perform a number of important functions in the cells of the human body. They play an important role in the tissues and organs of the human body in terms of construction, function, *etc*. They are made of monomeric units known as amino acids that are linked by peptide bonds. Proteins are also known as polypeptides [20 - 22]. Proteins are also hydrophilic in nature as they are made up of polar amino acid monomers. In a protein, several different amino acids may be linked together. Plant proteins are more immunogenic than animal proteins, have low molecular weight, and are mainly found in foods like corn, wheat, soybeans, *etc* [23].

Synthetic Biopolymer

Synthetic biopolymers are biological polymers that are either synthesized in the laboratory/industry or are prepared by modifying natural polymers. Synthetic biopolymers have several advantages over natural polymers in terms of their stability, and flexibility in various applications, and that is why synthetic biopolymers have been receiving lots of attention in the past decades. Moreover, synthetic biopolymers are preferable compared to synthetic polymers, as most of the synthetic biopolymers are biodegradable [23 - 25]. In recent years, synthetic biopolymers have been widely utilized in the medicine industry because of their specific properties like stability, controlled release, easy removal/degradability, and less or no immunogenicity. Synthetic biopolymers can be classified into the following two categories:

i. Biodegradable biopolymers and
ii. Non-biodegradable biopolymers.

Biodegradable polymers disintegrate by themselves due to the natural degradation through the action of microorganisms over some time. Due to their biodegradable nature, biopolymers become a viable alternative for packaging materials. Synthetic biodegradable polymers are more useful due to their mechanical properties. They also play an important role in tissue engineering. On the other hand, non-biodegradable biopolymers are a kind of biopolymer that cannot be broken down naturally by the action of microorganisms and end up as garbage, or a source of pollution [26 - 28]. They are mostly used in non-biomedical applications. The common examples are PA, PMMA, polycarbonate, PU, *etc*.

Classification of Smart Biopolymer

In the previous section, we discussed biopolymers and their classifications. Certain biopolymers tend to respond in the desired way due to small changes in pH, temperature, *etc*. known as smart biopolymers. In recent years, several smart biopolymeric materials have been developed as there is exponential growth in the application of smart biopolymers. Based on functionality and types of stimuli, smart biopolymers are classified into the following categories.

CHAPTER 10

Synthesis and Applications of Smart Polymer Nanocomposites in the Flexible Electronic Sensing

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Abstract: The use of smart electronic devices incorporated with sensors has become an indispensable part of the modern lifestyle. Such devices utilize the concept of flexible electronic sensing. The growing demand for flexible sensors is understandable because of their versatile application in wearable devices and other fields like robotics in the health sector. Continuous healthcare monitoring and personalized health concerns necessitate the development of flexible wearable devices. Flexible sensors play a crucial role in fabricating such wearable devices. In the realm of this, polymer nanocomposites (PNCs) have emerged as primary materials in flexible sensor design owing to their unique combination of electrical and mechanical properties *viz*. conductivity, mechanical properties, and chemical resistance. This book chapter explores the role of polymer nanocomposites in flexible sensor development. Additionally, various fabrication methods for sensor design with PNCs have been discussed. Moreover, recent advancements in utilizing PNCs for various sensor types are highlighted, paving the way for a new generation of comfortable and reliable wearable health monitoring devices.

Keywords: Core-shell structure, Diffusion-controlled, Drug delivery system, Flexible electronic, Magnetic hyperthermia, Magnetically-guided delivery, Mult stimuli-responsive, pH-responsive, Polymer-drug carriers, Smart polymers, Solvent-activated, Superparamagnetic iron oxide nanoparticles, Temperature responsive.

INTRODUCTION

In the last two decades, there has been rapid development in sensor-based technology owing to its versatile application in different areas [1 - 7]. In general, sensors can be classified into two broad categories based on their mechanical properties, *i.e.* rigid and flexible. Rigid sensors are usually made of low-cost

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materials, but they have limited flexibility. Due to their lack of flexibility, the application of rigid sensors is also quite limited. However, sensors for continuous health monitoring and other specific uses are required to be continually bent, stretched, or put under pressure, with a larger extent of flexibility [3, 7]. In this context, polymer nanocomposites (PNCs) have presented themselves as an excellent option for fabricating such sensors [7, 8]. PNCs are heterogeneous (multiphase) polymeric materials composed of at least one nano-scale phase (known as nanofiller) that is dispersed in a second phase (known as matrix) [9]. This heterogeneous blending provides PNCs with the combination of the individual properties of their constituents [9 - 11]. Owing to their excellent physio-chemical properties, they have been widely explored since their discovery in the early 1990s. The inherent flexible property of PNCs also allows for the creation of complex shapes, which can lead to reductions in the size, weight, and cost of the fabricated devices [7]. At present time, advancements in sensor technology have been achieved to develop flexible and biocompatible sensors based on PNCs [7, 8, 12, 13]. Such sensors can be integrated into wearable films, patches, and even temporary tattoos, *etc*. These devices utilise the concept of electronic sensing [14, 15]. Electronic sensing usually involves a sensor that responds to changes in the property being measured and converts it into an electrical signal [14]. The signal received can further be processed by suitable electronic circuits to interpret the data and provide an output that can be perceived by the users, *via* digital displays, analogue meters, or digital communication interfaces. The sensors continuously collect physiological data in real time, allowing for remote patient management by healthcare providers [1, 3, 16]. Machine learning algorithms enable healthcare professionals to optimize treatment based on the collected data from the sensors, assessing medication efficacy, safety, and recovery progress. These sensors are often referred to as electronic skin and are designed to mimic skin characteristics [3]. They can be placed directly on the human body to measure vital signs such as body temperature, heartbeat, and sweat composition. Advances in materials science and electronics have allowed for the development of electronic skin sensors with high resolution and fast response time while being flexible and stretchable [3]. Their flexibility makes them easier to integrate onto curved surfaces like the neck and wrist.

Considering the various applications of PNCs for the development of flexible sensors, in this chapter, we have highlighted the recent development of various types of flexible sensors. Various techniques used for the fabrication of flexible sensors with PNCs have also been discussed. With the advancement of material science and technology, sensors with enhanced flexibility, comfort, and responses are anticipated. Various applications of wearable sensors in the healthcare sector have been illustrated in Fig. (**1**) below.

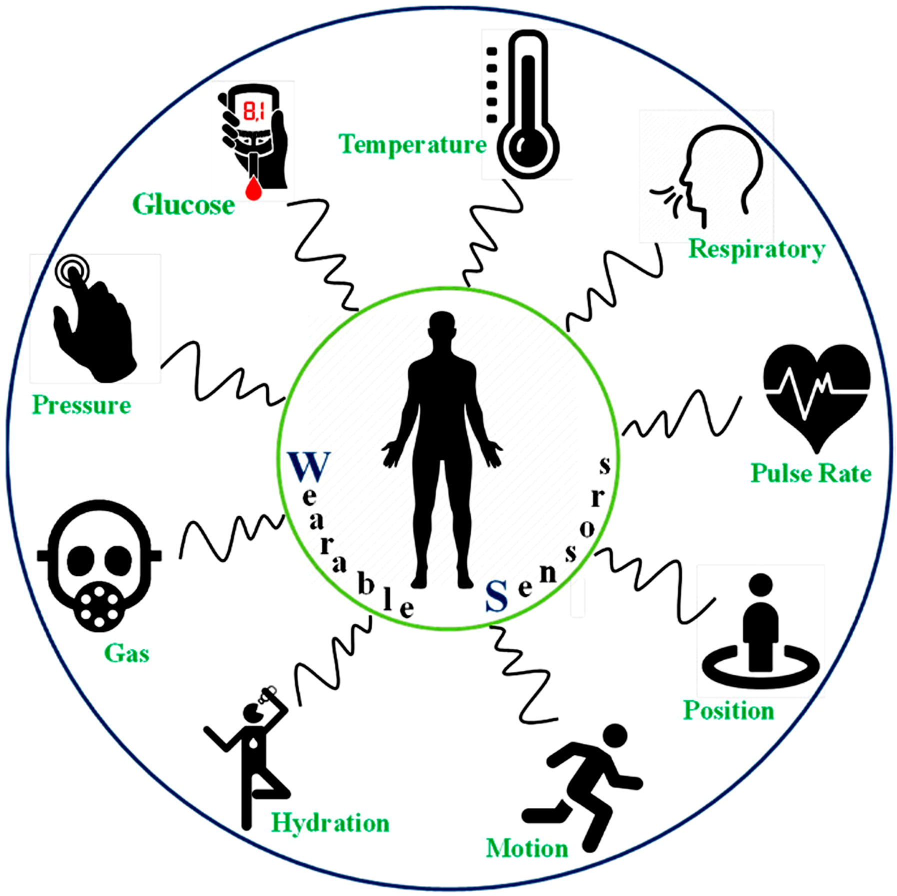

Fig. (1). Schematic of representative wearable healthcare sensing devices [16].

Material and Device Fabrication

The sensor's performance is influenced by the technique used for its fabrication [6, 17, 18]. Depending on the required resolution, sensor size and cost, flexible sensors are processed using various methods. Therefore, appropriate fabrication techniques are necessary for specific applications. For example, the Aerosol jet printing method is used for the fabrication of nano/microsensors at a very high resolution [6]. This is a 3D printing method which has been widely adopted for patterning high-resolution nanomaterial, thin film conductors, semiconductors, and dielectrics. However, as this method is not cost-effective, inkjet printing is used to print similar sensors with a lower resolution [6]. The inkjet printing technique is preferred for the large-scale fabrication of flexible electronic sensors. It is cost-effective because of its low material waste [19]. As a cost-effective and precise manufacturing method, inkjet printing is a versatile, controllable, and

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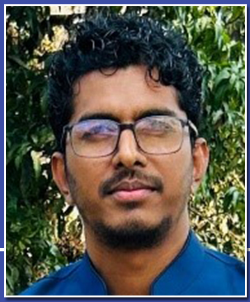
Partha Pratim Saikia

Dr. Partha Pratim Saikia is a distinguished academic and researcher with a background in organic chemistry, marked by a remarkable journey of academic achievements and contributions to scientific literature. He obtained his Ph.D. in Organic Chemistry from Dibrugarh University in 2011. Dr. Saikia is currently working as an Assistant Professor at N.N. Saikia College, Titabar, Jorhat, since 2012. He has been recognized for his outstanding contributions, earning prestigious awards such as the "Most Cited Paper Award" by Elsevier for consecutive periods, as well as Junior and Senior Research Fellowships from the Council of Scientific and Industrial Research, India. His research projects have been supported by renowned funding agencies, exemplified by his ongoing DST-TARE fellowship, which focuses on the extraction of electrical energy from the hydrological cycle. Furthermore, Dr. Saikia has contributed to the scientific community through 30 articles in international journals and 5 chapters published in esteemed books, and he has filed multiple patents for innovative chemical compounds and processes.



Bhaskar Jyoti Saikia

Dr. Bhaskar Jyoti Saikia is a highly accomplished researcher and scholar in the field of polymer chemistry. He obtained his Ph.D. from Tezpur University for his work on the synthesis of functional polymers through Atom Transfer Radical Polymerization (ATRP) and their fluorescence and self-healing characteristics. Dr. Saikia is currently working as an Assistant Professor at N.N. Saikia College, Titabar, India, since 2016. His research work has earned recognition and accolades, including prestigious awards such as the 7th National Award for Technology Innovation in Petrochemicals & Downstream Plastics Processing Industry by the Ministry of Chemicals & Fertilizers, Government of India. Additionally, he has been honored with the Dr. Santi Nath Ghosh Memorial Award and the Conference Merit Award for his outstanding presentations at the national level, highlighting his scholarly excellence and innovative research.

His research contributions are both prolific and impactful, with 20 publications in reputable international journals and 5 book chapters covering a wide range of topics, from the synthesis of smart polymers to the development of nanocomposites with diverse applications. Dr. Saikia's work demonstrates his expertise in designing novel materials with tailored properties, as evident from his studies on self-healing polymers, responsive nanocomposites, and electroactive materials for energy storage devices.