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SMART BIOCOMPOSITE MATERIALS

FABRICATION, APPLICATIONS, AND SUSTAINABILITY

Edited by

MD. REZAUR RAHMAN

MUHAMMAD KHUSAIRY BIN BAKRI

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Smart Biocomposite Materials

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Dedication

This work is dedicated to my amazing wife and daughters, Shirin Akther, Fahriah Rahman, and Faizah Rahman, who are very special to me and made it possible for me to complete this work.

—Associate Professor Ts. Dr. Md. Rezaur Rahman

First, I would like to thank the Almighty God for the guidance, strength, power of mind, protection, and for giving us a healthy life. All of these we offer to you. Every difficult task needs self-effort as well as the guidance of elders, particularly those who are near to our hearts. I offer my humble dedications to my beautiful and loving father, mother, wife, and brothers, whose devotion, love, support, and nightly prayers have enabled me to work toward this significant achievement, along with all the dedicated, well-liked, and well-respected teachers and supervisors.

—Ts. Dr. Hj. Muhammad Khusairy Bin Bakri

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Preface

Materials science has now reached a critical phase of development, where combining sustainability, intelligence, and functionality is no longer visionary but the absolute requirement of the time. While the world is facing increasingly aggravated environmental concerns and its industries face growing demands for responsive and high-performance materials, the combination of biobased components with intelligent attributes is an opportunity for innovation, at the same time as it is eco-friendly and technologically advanced.

Intelligent biocomposite materials are one class of engineered systems combining renewable natural fibers with biodegradable or recyclable matrices and functional fillers or additives with responsive behaviors to external stimuli. These materials successfully address the imperative of reducing dependency on fossil-derived resources and possess enhanced features like self-healing, shape memory, and active sensing or actuation functions as well. The last 10 years have seen tremendous development in this field, driven by interdisciplinarity among materials engineering, biotechnology, polymer science, nanotechnology, and applied mechanics.

At the core of this discussion is a deep understanding of the interactions between different components, natural fibers, polymer matrices, and added functionalities. Advances in fabrication technologies, ranging from conventional compounding methods to advanced additive manufacturing methods, have enabled the large-scale production of biocomposites with tailored mechanical, thermal, and functional properties. The combination of responsive functionalities, such as sensors, stimuli-responsive fillers, or drug delivery systems, greatly expands their utility in high-end markets, such as biomedical devices, aerospace components, structural health monitoring, and soft robotics.

An important aspect of research in this area is mirroring the built-in self-sufficiency of nature with the development of materials with self-healing tendencies, the ability to change their shape in the presence of outside stimuli, and the ability to perform regulated-release functions. These attributes improve the potential of biocomposites and open up new prospects for research and application.

Sustainability remains the underlying tenet of this field, including not just the origin of materials but also life cycle assessment, recyclability, and environmental impact considerations. As the world economy increasingly moves toward more circular and biobased models of development, novel biocomposites are expected to play a key role in enabling this transformational process.

This book aims to act as a comprehensive scholarly reference for researchers, engineers, and postgraduate instructors to explore the scientific basis, processing methodologies, functional characteristics, and potential uses of intelligent biocomposite materials. This book integrates fundamental knowledge and state-of-the-art advancements to prompt further research and enable the development of advanced materials with features like intelligence, long-term performance, and sustainability.

Associate Professor Ts. Dr. Md. Rezaur Rahman
Ts. Dr. Hj. Muhammad Khusairy Bin Capt. Hj. Bakri

Applications of biocomposites in biomedical engineering

11

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11.1 Introduction

Biocomposite is the combination of two or more distinct constituent materials which one of it is a naturally derived based materials such as the combination of biopolymers and reinforcing agents to overcome the issue or lacking in one individual constituent materials and creating a new and novel material foe various purposes (Al Mahmud, 2023; Rudin et al., 2013). This type of material has been widely investigated to apply and improve the existing biomedical engineering field due to its unique ability and characteristics that are highly biocompatible with designed properties based on its application (Al Mahmud, 2023). A multifunction biocomposite is a combination of biocomposite materials with biodegradability as a continuous phase owing to its processability and shape stability and bioactive agents as filler that contribute the composite with multifunction such as immunomodulatory function and antibacterial function (Gao et al., 2021; He et al., 2017; Ou et al., 2020). This type of biocomposite that is integrated into the scaffold could introduce an antibacterial and immunomodulation environment to the defect area, which promotes higher osteogenesis ability within the scaffold and results in a better scaffold fabrication option (Adamu et al., 2019; Bakri et al., 2018; Fatema et al., 2024; Hari et al., 2021; James et al., 2024; Jayamani et al., 2015, 2016).

On top of that, a multifunctional biocomposite could be further improved by introducing a stimuli-responsive biomaterial into the composite, which denotes it as a smart biocomposite. Designing smart and adaptable biocomposite materials that change their properties depending on the external stimuli within a controlled and reversible action has gained significant interest in the biomedical engineering field (Adam et al., 2023). A stimuli-responsive biomaterial is the main component in fabricating a stimuli-responsive scaffold in tissue engineering, a type of scaffold that is able to control stem cells' behavior precisely as required for the regeneration and restoration of organs. A stimuli-responsive biomaterial scaffold was stated to have the ability to imitate the endogenous signals that influence the extracellular microenvironment of stem cells toward a desired environment via a precise control of exogenous stimuli, which provides a more advanced and versatile approach to cellular behavior manipulation (Gelmi & Schutt, 2021).

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A smart hydrogel has been widely studied and applied in various field such as designing a biomedical material, industrial material, agricultural material, electrical material, healthcare and hygiene products, also utilization in cell adhesion application, self-healing wound dressing, drug delivery devices, wearable electronics, tissue engineering, energy storing materials, and wastewater management owing to its excellent responded characteristics toward various environmental conditions, for instance, pH, temperature, light, electric and magnetic field, biological stimuli, and chemical stimuli (Gao et al., 2019; Ravishankar et al., 2019; Sikdar et al., 2021; Tang et al., 2020). Hydrogel is a material that has tunable physicochemical properties, particularly its mechanical properties, elasticity, and viscoelasticity, where it could provide specific functions toward its application, such as the stem cell behavior and fate (Ma et al., 2018, 2021). The addition of stimuli-responsive ability to the hydrogel causes it to have controlled and adaptable properties that are easily applied to various applications, especially in biomedical cases.

The swelling properties of the hydrogel play a significant role in influencing the stimulus responses of a modified smart hydrogel in biomedical applications. In a drug delivery system, the swelling behavior of the hydrogels was stated to determine the effectiveness of the hydrogel in sustaining the release of the drugs in the targeted area (Ali et al., 2023). A chitosan hydrogel was found to have the ability to swell or collapse depending on the pH and ionic concentration of the changes in the external environment (Ma et al., 2022). Hence, a smart biocomposite hydrogel with incorporation of various materials would be further discussed in this chapter, where the purpose of the smart biocomposite hydrogel in tissue engineering, wound healing management system, drug delivery system, and cancer and tumor therapeutic applications is the main focus of this chapter. This chapter also covered the role and method in which pH-responsive, temperature-responsive, and magnetic-responsive materials act in the smart biocomposite system in the biomedical engineering application field. The challenges and issues with future roads that other researchers could take on were also discussed at the end of this chapter.

11.2 Application of smart biocomposites in tissue engineering

Tissue engineering is a field that aims to repair, replace, and restore damaged or diseased tissue and generate healthy tissue by combining cells, scaffolds, and growth factors (Han et al., 2020). The development in this field has provide surgeon with a varied choice in tissue restoration especially in bone tissue restoration form and function as this alternative could assist in the bone tissue cell migration, proliferation and differentiation by providing a cell friendly microenvironment with three main factors that are the cells, scaffolds, and growth factors (Henkel et al., 2013). Fig. 11.1 shows the general process involved in tissue engineering, where the fabrication of a scaffold with growth factor and preseeded cells is the starting step followed by the scaffold implantation into the individual system and cell infiltration, vascularization, and neo-tissue formation to restore and regenerate the intended organ tissue. In bone

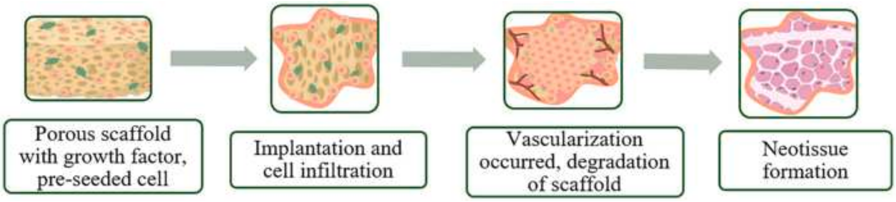

Figure 11.1 The schematic diagram of the process involved in tissue engineering. The process involved four steps for repairing, replacing, and restoring the damaged tissue organ into a healthy tissue.

tissue engineering, a material that can be instituted to the bone defect section in the patient recipient's system and remodeled by the individual's cell after the implementation process with lower chances of further surgery required is the main objective of this field (Koons et al., 2020; Shaw et al., 2018).

Therefore biocomposites have been closely associated with tissue engineering as a material for the scaffold fabrication, and their properties have a higher impact on the efficiency of the scaffold in substituting the extracellular matrix of the bone in the bone defect area. An ideal biocomposite and biomaterial should have highly biocompatible and biodegradable properties, which will not trigger any chronic immune response and could be replaced by the patient's bone at a similar rate of new bone formation (Perić Kačarević et al., 2020). A smart biocomposite and biomaterial has gained recognition in the biomedical field as it can provide a tunable material depending on the biomedical cases, as this material can change its properties into a desirable outcome when exposed to external forces that could be precisely controlled. The magnitude of the applied exogenous stimuli signal influences the limitation of the biocomposite response (Gelmi & Schutt, 2021).

11.2.1 Scaffold purpose in tissue engineering

One of the main goals in tissue engineering is designing and creating an artificial scaffold that has the potential to substitute a damaged tissue (Doshisha University, 2024). This is due to the significant role it provides in tissue engineering as a platform for the attachment of preseeded cells and exogenous growth factors for promoting neo-tissue formation (Pfau & Grunlan, 2021). In bone tissue engineering, an artificial bone scaffold has become an ideal technique for bone defect repair as it has good biocompatibility, degradability by-products, and accessible sourcing since most of these scaffolds are made from synthetic polymers, natural polymers, and inorganic materials (Hu, Yang, et al., 2024). Besides, they have also concluded that a porous microsphere scaffold is an essential feature of a good scaffold to ensure the effectiveness of osteoconductive properties, which influence the growth of bone tissue, such as the fibrous tissue within the scaffolding microsphere. This finding is supported by previous research, where they also highlighted the importance of osteoconductivity, osteoinductivity, biocompatibility, and mechanical strength that matched the natural tissue factor for an implantable scaffold (Leppik et al., 2018; Mallick et al., 2019; Wei et al., 2022).

It has been noted that the irregularity of bond defect size after surgical debridement and various bone tissue morphology, based on its location, has increased the needs of an artificial bone scaffold with customizable shapes, which leads to the introduction of 3D printing technology and hydrogel injection in scaffold fabrication methods (Hu, Chen, et al., 2024; Hu, Yang, et al., 2024). Hu, Yang, et al. (2024) emphasized the importance of pore structure and mechanical attributes factor in scaffold fabrication, especially for artificial bone scaffolds. Additionally, a scaffold that has been customized with mechanical properties and degradation profile strives to support the mechanical characteristics of an actual bone and match the rate of tissue formation in biomedical applications (Pfau & Grunlan, 2021). Hence, a smart biocomposite scaffold has been introduced and rapidly studied as an alternative material for tissue regeneration, repair, and restoration.

11.2.2 Temperature-responsive shape memory scaffold in tissue engineering

Shape memory scaffold is one of a smart artificial scaffold that utilized a shape memory polymer, which is a type of smart material that could changes its shape by setting it to remember a temporary shape and revert into its original shape under external stimuli such as temperature and light (Chen et al., 2024; Pfau & Grunlan, 2021). Pfau and Grunlan (2021) affirmed that the response, which is also known as the switching phase or segments, consists of shape fixity, a process of deformation followed by securing into a temporary shape, and shape recovery, where it returns to the original permanent shape. The temperature-responsive shape memory scaffold holds a significant benefit in reducing the difficulty and trauma of a surgery as it requires the smallest invasive implantation condition (Bian et al., 2023; Hu, Yang, et al., 2024). Furthermore, the natural human body temperature was utilized in a temperature-responsive shape memory scaffold to obtain an instant shape recovery, which gains attention in tissue engineering (Hu, Yang, et al., 2024).

Hydrogel is a type of material that is made up of a hydrophilic polymer network and can absorb water or biological fluid without dissolving, which makes it a highly biocompatible material (Hillel et al., 2007; Thang et al., 2023). Additionally, the flexibility in changing its degree of swelling in response to external stimuli such as temperature has rendered it a shape memory polymer in biomedical applications (Laftah et al., 2011). Thus various thermo-responsive hydrogels have been studied to be implemented as a biocomposite material or a fixed shape/memory biocomposite material for scaffold fabrication in tissue engineering, such as poly(*N*-isopropylacrylamide) (PNIPAM), poly(*N*-acryloylglycinamide) (PNAGAm), polyurethane, and polylactic acid (PLLA) (Adam et al., 2023; Chen et al., 2024; Hu, Chen, et al., 2024; Nishimura et al., 2024).

A thermo-responsive PNIPAM is a type of temperature-sensitive hydrogel that is flexible within a varied range of temperatures, particularly in the range of human body temperature, in which the response of PNIPAM depends on the lower critical solution temperature (LCST) resulting in volume phase transition of the material (Adam et al., 2023; Heskins & Guillet, 1968; Hirokawa & Tanaka, 1984; Islam et al., 2014).

It was stated that when the temperature is below the LCST, the hydrophilic amide groups of PNIPAM will form hydrogen bonds with the surrounding water molecules and trap them in the structure. Then, by increasing the temperature above the LCST, the trapped water from the structure is released as the hydrophobic interaction between the propyl groups of PNIPAM is highly favored, resulting in the polymer chain breakdown process. Additionally, a hydrogel consists of PNAGAm, an amino–acid–derived vinyl polymer that has strong hydrogen bonds side chains between the polymer chains, has the same ability as PNIPAM, but its volume phase transition depends on the upper critical solution temperature (Majstorović et al., 2023; Nishimura et al., 2024). The volume phase transition ability of the hydrogel, in which the polymer bonds breakdown at a high temperature and reattach at lower temperature, gives a unique ability to the polymer through remembering and recovering its shape in the response toward the changes of temperature (Jayamaui et al., 2020; Kiew et al., 2013; Kuok et al., 2024; Lai et al., 2015; Rahman et al., 2011, 2017, 2019; Rahman, Hamdan, Hasan, et al., 2015; Rahman, Hamdan, Hashim, et al., 2015).

Despite their fixed shape/memory properties, these hydrogels have low mechanical strength if applied as is without alteration, which limits their application in various fields due to their degree of swelling ratio of the particles. Adam et al. (2023) have stated that a highly swollen hydrogel caused the polymer chains to become highly flexible that resulting in lower mechanical strength. Nevertheless, a study has also found that the higher swelling ability of the polymer improved the tolerability of these hydrogels in changing its shape, but in terms of its healing ability, a lower swelling factor is highly favored due to the higher PNAGAm molecules mobility in the hydrogel when the swelling factor is lowered (Majstorović et al., 2023). Therefore, depending on the hydrogel applications, the effect of the swelling factor could impact the hydrogel's ability and benefits. Various researchers have addressed these issues by implementing the hydrogel in a suitable composite system with its desired properties.

Research has focused on the effects of thermal-sensitive hydrogel mechanical properties toward a collagen network architecture-dependent mechanics by embedding thermo-responsive PNIPAM microgel particles into the collagen network via the shear rheology method (Adam et al., 2023). They have concluded that the combination effect of collagen network architecture and the initial polymerization state of the biocomposites influences the switching response of the hydrogel polymer and collagen network from hydrophilic to hydrophobic nature or vice versa in reliance on the composite's polymerization temperature. This situation caused an improvement in the elastic modulus of the materials as the polymerization temperature varied to 25°C and 37°C, and it was recommended to be applied as a new diagnostic and therapeutic application in the tissue engineering field (Adam et al., 2023). Moreover, Nishimura et al. (2024) have investigated the relationship between the elastic modulus of hydrogels and the cell behavior in the application of tissue scaffold fabrication, where they copolymerized *N*-acryloylglycinamide (base polymer) with polymerizable arginine–glycine–aspartic acid-serine peptide (cell-binding site) without chemical crosslinker. This chapter has successfully designed a hydrogel system that has adjustable elastic modulus via compression in various thicknesses at high temperatures, as the hydrogen bonds within the polymer were

Figure 11.2 Illustration of shape memory scaffold process. The process of a thermo-responsive shape memory scaffold implementation for an irregular bone defect area by PLLA-TMC-GA and PHA scaffold.

broken and reattached after it was exposed to a lower temperature, causing it to maintain the shape and elastic modulus.

Besides that, a linear relationship between the elastic modulus and cell adhesion behavior has been discovered, which depicts the ability to utilize the shape memory properties of hydrogel as cell growth factors (Nishimura et al., 2024). Furthermore, the utilization of thermo-responsive shape memory scaffold in irregular bone tissue defect treatment has been studied in which they have successfully proven the ability of PLLA-trimethylene carbonate-poly(glycolic acid) encapsulated by dopamine-modified-nanohydroxyapatite (PHA) with the addition of icariin scaffold to provide a shape memory function with high mechanical strength and positive response of transforming to biological bone within the duration of human bone natural regeneration, along with biodegradability by-products produced (Hu, Yang, et al., 2024). They have also suggested a method for the multitiered microsphere scaffold implementation process that guarantees a close contact between the scaffold and peripheral bone defect during surgery, as shown in Fig. 11.2.

11.2.3 Stimuli-responsive scaffold in tissue engineering

Stimuli-responsive scaffold is a type of smart scaffold that has the potential to independently respond to specific environmental changes surrounding the affected area which resulting in improvement of bone defect regeneration by the scaffold (Wei et al., 2022). The changes in pH reduction and enzymes due to the secretion of bacteria in a severe infection area with an excess reactive oxygen species (ROS) or a mild acidity environment in tumor areas are examples of specific environmental changes (Chen et al., 2021; Deng et al., 2020; Dong et al., 2020; Pourhajibagher et al., 2020). Metal-phenolic networks (MPNs), a type of supermolecular complex that is made from a combination of phenolic ligand (tannic acid [TA]) and metal ions through coordination bonds, have served as pH-responsive materials owing to their disassembly kinetics as a capsule in drug delivery and a filler for multifunctional biocomposites (Gao et al., 2021; Guo et al., 2014). The advantages of utilizing MPNs are the combination of phenolic ligands and metal ions that give a combination of each perspective biofunction (Min et al., 2022). On top of that, varying the metal ions while maintaining the type of phenolic ligands could increase the ability and function of the biocomposite scaffold, which provides more options for the scaffold implantation depending on the defect area requirements.

TA has been successfully integrated as the phenolic ligand while magnesium ions, Mg^{2+}, as the metal ions for MPNs filler in a polycaprolactone (PCL) nanofibrous scaffold where a pH-responsive release action was detected along with an improvement on the osteogenic performance through vivo analysis (Gao et al., 2021). They have recorded that the presence of a pH-responsive release action of the TA–Mg^{2+}-based MPNs filler has introduced an osteoimmune microenvironment to the surrounding area, which concurrently increased the cell adhesion and stimulated osteogenic differentiation of the stem cells in the PCL nanofibrous scaffold. On the other hand, another study has successfully integrated the effect of zinc metal ion (Zn^{2+} ions) as an antibacteria component for a TA/Zn^{2+}-based magnetic nanoparticles (MNPs) filler in a PCL nanofibers scaffolds with the aim of comprising a filler with immunomodulatory functions through antibacterial components and can regulate a long-term release behavior of these components (Min et al., 2022). They have concluded that the ability of the filler to regulate the initial acute inflammation at a moderate level and fully impede the chronic inflammation induced at the later degradation stage of the modified scaffold. Thus, varying the metal ions could introduce new functions to the modified scaffold for the tissue regeneration process in the tissue engineering field, particularly for bone tissue engineering. Nevertheless, the release behavior of the MNPs needs to be considered when selecting the phenolic ligand and metal ions, as each metal ion has a different affinity toward the phenolic ligands (Min et al., 2022).

11.3 Application of smart biocomposites as wound healing management

The role of smart biocomposites in wound healing management usually centers on the pH-responsive biomaterial, such as a pH-responsive hydrogel biocomposite and pH-responsive membrane biocomposite. The pH of a wound area and wound fluids serve as an essential factor in wound healing protocol, in which it could be varied depending on the types of wounds, the phase of the wounds, and the presence of infection bacteria (Bowler et al., 2001; Dhivya et al., 2015). Wound care is important to prevent complications in human lives that could lead to death if the wound area is exposed to a serious wound infection. The misuse and overuse of antibiotics, antidotes, new drugs, unsuitable pH levels for a certain type of wound, and inadequate wound dressing were examples of causes for wound infection (Ren et al., 2020).

A bacterial infection has posed a serious issue in wound healing after surgery since it disrupts the healing process through the continuous inflammatory process and has been treated through the application of antibiotic agents (Luo et al., 2020). It has been discovered that the pathogenic bacteria such as *Escherichia coli*, *Staphylococcus* aureus, and *Pseudomonas aeruginosa* have high resistance toward antibiotics due to the misuse of antibiotics, which greatly affects human beings (Caniça et al., 2019; Domingues et al., 2021; Gupta et al., 2019). This problem has encouraged researchers to construct alternative methods or new antibacterial components to be used in a postoperative wound healing and drug delivery system. Recent studies have focused on nanotechnology utilization (Namakka et al., 2023; Rahman et al., 2024; Sueraya et al., 2024), such as metal-organic frameworks (MOFs), as a potential antibacterial agent. Luo et al. (2020) have studied the antibacterial and antiinflammatory factor of a zeolitic imidazolate framework-8 (ZIF-8) as an MOF with sodium alginate-niflumic acid biocomposite hydrogels for a local wound infection treatment. Additionally, ZIF-8 has also been implemented together with silver nanoparticles (Ag)-physcion (Phy)-hyaluronic acid (HA) biocomposite for introducing an antibacterial effect with controllable release of the component, especially Ag NPs on infectable wound area (Tan et al., 2022).

They have highlighted the unique characteristics of ZIF-8, a type of MOF structure that has been widely implemented in biomedical applications. MOFs are 3D porous materials with high porosity and surface area, high stability, modifiable structure, and excellent biocompatibility, which has been reported to hinder bacterial colonization and biofilm formation, making it an excellent antibacterial dressing (Luo et al., 2020; Tan et al., 2022). ZIF-8 is the most biocompatible MOF structure with unique pore size and surface area, chemically stable and has a pH-responsive ability that could help in controlling the release rate of the drugs (Luo et al., 2020; Tan et al., 2022; Wang, Sun, et al., 2020). A study has discussed the pH release mechanism of the Ag-Phy-HA-ZIF-8 biocomposite that disintegrated under acidic conditions, resulting in the release value of Phy and Ag^+ being 2.5 times and 3 times higher in acidic conditions, respectively (Tan et al., 2022). It was noted in the paper that the existence of bacteria favored in a micro-acidic environment, which proved the smart release behavior of an antibacterial factor in the biocomposites to the infected area only and had no effect on the healthy area.

Besides that, wound care management also plays a significant role in wound dressing, healing rate, and protection against infections. The pH of a typical type of wound after surgery is usually of a basic nature that ranging around 8.5, and is gradually reduced as the wound starts its healing process (Bennison et al., 2017; Ren et al., 2020). An antimicrobial action of pH-responsive TA-keratin (KA) hydrogel biocomposites cross-linked with high surface area of graphene oxide quantum dots has proven in its perfect healing ability of a wound area within a shortest duration without subsequent infection and no visible hard scab observed (Ren et al., 2020). In this paper, the modified hydrogel biocomposite shows a higher swelling percentage at pH 10 and lower swelling percentage at pH 1 and pH 3, which depicts the diffusion and the release of the drug content in TA-KA hydrogel at basic conditions since TA-KA hydrogel is composed of acidic groups that will swell quickly in basic solutions via deprotonation and repulsion action within the acidic chain polymers. Thus it was recorded to be appropriate material for wound healing applications.

Other than that, a biocomposite hydrogel that aims toward diabetic wound healing has also been investigated recently, since a diabetic wound has weak immunization and has a high chance of being exposed to bacterial infection as well as inflammatory reactions. Various past papers have noted that the hyperglycemia and weak immunity of diabetic wound patients, in addition to the complexity and difficulty of the wound type, are the causes for the diabetic wound to be infected and lead to a delay in the wound healing process or worse, an amputation of the infected area (Atamna et al., 2019; Hu et al., 2022; Tu et al., 2022; Vijayakumar et al., 2019). The difficulty in managing a diabetic wound was because of the reoccurrence of the inflammatory response, oxidative stress, and bacterial infection which has underlined the significant of a timely monitoring toward the changes in the diabetics wound so that information and timely action could be executed instantly to prevent a chronic state of the wound (Moura et al., 2013; Xie et al., 2023; Zhang et al., 2022).

Thus a study for enhancing the diabetic wound healing by monitoring the wound area with pH-sensitivity factor has been conducted where they deployed a multifunctional poly(γ-glutamic acid) hydrogel encapsulated with KA-based hydrogen sulfide (H_2S) donor, ciprofloxacin, and anthocyanins into a full thickness cutaneous diabetic wound model in rats (Liu et al., 2023). They have utilized a color-changing wound dressing as a platform for timely detecting a bacterial infection and making adjustments to the treatment via natural dye indicator, anthocyanins in the modified biocomposite hydrogel that produces a marker based on the pH conditions of the wound microenvironment. Bennison et al. (2017) have stated that the pH value for healthy skin ranges from 4 to 6.3, which is slightly acidic and will increase to become more alkaline if it becomes an infected wound. Based on these indicators, the clinician could timely diagnose the wound microenvironment and address the suitable direction for diabetic wound treatment or any type of wound.

11.4 Utilization of smart biocomposites in drug delivery systems

Drug delivery system is a device and formulation that is programmed to deliver therapeutic substances into the body in a more efficient, safe, and stronger medication potency delivery method with a controlled release ability via organic and inorganic structures carriers (Jain, 2020; Nabipour et al., 2024). Inorganic and organic carriers may have toxicity with low degradability and low leading capacity respectively (Wang, Yan, et al., 2020). A pH-responsive drug delivery system as a controlled drug release method that could remain in the therapeutic range for an extended period under a regulated time and drug release area has garnered considerable interest in the drug delivery system field (Joo et al., 2013; Zangeneh et al., 2022; Zeraatpishe et al., 2019). Thus a smart biocomposite is applied to the drug delivery system as a carrier for administering therapeutic medicine in the human body for various types of disease and cases, such as epilepsy therapy drugs, aphthous ulcer cases, and antidepressant drugs.

A pH-responsive nanobiocomposite has been studied as a drug carrier material that has the ability to restrain a phenytoin sodium (PHT) release in a burst pattern through a halloysite nanotubes (HNTs) surface modification with zein protein and a layer-by-layer technique (Zangeneh et al., 2022). PHT is an anticonvulsant drug that is extensively applied in epilepsy therapy, a type of chronic neurological disorder with nerve attacks in seizure form (Haut et al., 2006). In this paper, they have emphasized that the pH release medium and zein polymeric coating was acted as a barrier to the

PHT burst release action, in which the factor for a controlled drug release manner in the system concurrently shows the ability of this drug carrier to protect the drug in the stomach environment. Additionally, they have explained that zein polymer can release acidic hydrogen in an alkaline environment and obtain a negative charge that breaks the polymer chain, which then releases PHT into the area.

The pH-response of the hydrogel was found to be the same as in research conducted by Ali et al. (2023), where they recorded a high degree of swelling for pH-sensitive copolymeric hydrogel based on Salvia spinosa seeds (SSH) and acrylic acid (AA) with inclusion of venlafaxine hydrochloride (venlafaxine HCL), an antidepression drug in an alkaline environment with pH 7.4 and human body temperature of 37°C. It was stated that this was due to the COOH groups in SSH-co-AA hydrogel that were initiated to ionize into an anionic form that caused an electrostatic repulsion (anion–anion) within the polymer chain, which then broke up the polymer chain and increased the swelling behavior. On the other hand, they also portrayed the dependence of venlafaxine HCL release rate on the swelling behavior of the hydrogel. Thus, they concluded that strengthening the hydrogen bonds and development of physical interaction between the hydrogels was the key factor in influencing the swelling behavior of the hydrogel, which directly affects the pH response in drug release.

On the other hand, a study in which a gelatin based mucoadhesive hydrogel biocomposite was found to be releasing the drug substances more in an acidic environment as the swelling of the modified hydrogel increased, due to the favorable condition of the hydrogen bonding interaction in the hydrogel active functional groups (Ramezanalizadeh & Delgoshaee, 2024). This paper studied the pH-responsive of a gelatin/EudragitL100/Xanthan gum with inclusion of titanium oxide (TiO_2) and copper oxide (CuO) metal ions as a gelatin based mucoadhesive hydrogel for treating aphthous ulcer disease, as successful mucoadhesion drug delivery materials with antibacterial and biocompatibility properties, a positive cell transport and adhesion activity and an easy application approach. The contrast result obtained in Ramezanalizadeh and Delgoshaee (2024) in comparison to Ali et al. (2023) and Zangeneh et al. (2022) has shown that the relationship between the hydrogel component and the strength of hydrogen bonds is important for a pH-responsive hydrogel biocomposite, depending on the purpose of the drug that they applied. Since Ramezanalizadeh and Delgoshaee (2024) were focusing on the release of the drug toward aphthous ulcer disease that was due to stomach acidic condition, the swelling of the hydrogel needed to be risen in acidic environment while the other two studies were focusing on delivery of the drugs to the small intestine for it to be absorbed into the body.

11.5 Role of smart biocomposites in cancer and tumor therapeutic applications

11.5.1 Controlled anticancer drug release and cancer vaccines

Recent studies have utilized the unique ability of MOFs and MPNs in an anticancer drug delivery system. The ability of designing various functional materials with sufficient porosity and adjustable pore size for MOFs fabrication, while the ability of MPNs structure through a combination of a phenolic ligand, that has an antiinflammatory function which target robust reactive oxide species (ROS) and metal ions as the vital enzymes, and transcription factors were the key focus of various studies on their role as anticancer drug carriers (Li et al., 2023; Min et al., 2022; Nabipour et al., 2024). A TA/Zn^{2+}-based MNPs filler has been designed and implemented in PCL nanofiber scaffolds that can reduce and inhibit an acute or chronic inflammation in the defected area with robust ROS scavenging ability, in addition to a pH-response toward the release of Zn^{2+} ions that indirectly prevent tumor genesis (Min et al., 2022).

Moreover, in a study on carboxymethyl cellulose (CMC)/bio-based copper-based MOF (bio-MOF) hydrogel bead with encapsulation of curcumin (CUR), an anticancer drug that exhibited antiinflammatory, antioxidant, antibacterial, and anticancer properties, was deployed to cervical cancer and neuroblastoma cell lines to develop a bio-based carrier that has a controlled and pH-sensitive ability in releasing CUR to a targeted area (Nabipour et al., 2024). They have demonstrated the effectiveness of the CMC/CUR-bio-based MOF biocomposite hydrogel in delivering a controlled release of CUR with pH-sensitivity, under physical conditions and acidic environment of tumor cells, which ares pH 7.5 and pH 5.0, respectively. Additionally, they also concluded that a slowed and lowered release of CUR drug at pH 5.0 depicts the advantages of the modified hydrogel for targeting low pH intracellular endosomes and lysosomes, thus improving the cytotoxicity of these biocomposites toward the cancer tissues. In this paper, the ability of the CMC/CUR-bio based MOF biocomposite hydrogel to release a minimum amount of drug leakage under pH 7.0 was highlighted, which reduces the adverse effect of the drug on normal tissue around the targeted area.

Besides that, another research has focused on reducing the side effects of chemotherapy drugs such as doxorubicin (DOX) under a controlled release manner and overcoming the multidrug resistance of tumor cells in which a poly(acrylic acid-*co*-2-hydroxyethyl methacrylate)/rutin hydrogel was developed as a drug carrier of DOX while utilizing a pH-responsive polymer to regulate the drug release substances in a controlled manner (Onder et al., 2023). They have explained that the amine groups in DOX were protonated under an acidic environment which result in an increment of hydrophilic nature and higher solubility, which concurrently releases the DOX. The electrostatic interaction between the positively charged DOX and the negatively charged carboxylic groups in the AA that protonate in a lower pH environment, has caused the DOX to be released in a faster way. The inclusion of rutin, a natural polyphenol that has the ability of making the cancer cell susceptible to chemotherapy drugs by introducing a flow pump inhibition, apoptosis activation, and immobilization of the cell cycle toward the cancer cell, into the biocomposite hydrogel has increased the efficiency DOX in apprehending the cancer cell lines, such as a triple negative breast cancer within a low dose has been proven in this paper (Iriti et al., 2017; Onder et al., 2023).

11.5.2 Magnetic nanoparticles in hyperthermia cancer therapy

Hyperthermia cancer therapy is a cancer treatment that is also known as a thermal therapy technique that involves temperature and magnetic properties from MNPs to destroy a malignant tumor by heating the surrounding area to a temperature above the normal conditions, ranging from 41°C to 45°C within a certain period (Bahojb Noruzi et al., 2024; Eivazzadeh-Keihan et al., 2022). MNPs are magnetic metal oxides with a particle size of 1–100 nm and exhibit special characteristics that attract researchers to utilize them as the heating medium in hyperthermia therapy (Bahojb Noruzi et al., 2024). The MNPs were stated to have a high-surface-to-volume ratio and the ability of easily separated using an external magnet, also generating local heat when it was placed in an alternating magnetic field (AMF). Iron oxide, $Fe_3O_{2,}$ is the most widely used magnetic nanomaterial in biomedical applications, especially in hyperthermia treatment due to its nontoxic nature, antibacterial properties, superparamagnetic actions, and high biocompatible with the human body (Bahojb Noruzi et al., 2024; Eivazzadeh-Keihan et al., 2021, 2022). This cancer treatment method has low damaging effects on the surrounding healthy tissue as it selectively targets the cancer cells, which depicts its favorable approach as a cancer treatment other than chemotherapy and radiotherapy (Pajoum et al., 2024).

The hyperthermia cancer treatment process starts with the injection of the MNPs that were in a suspension of liquid carrier into the tumor area via direct injection or into an artery that is directed toward the tumor (Bahojb Noruzi et al., 2024; Eivazzadeh-Keihan et al., 2022). Once the MNPs have been inserted into the tumor area, the AMF is used to heat them, in which magnetic energy is transferred into thermal energy, and this thermal energy eventually spreads out to the surrounding tumor tissue without damaging the healthy tissue through a controlled and precise measured. Bahojb Noruzi et al. (2024) have emphasized the important factors of the MNPs size, shape, magnetic properties, and the magnetic field strength and frequency, which influence the amount of heat transfer to the tumor area. Specific absorption rate (SAR) is the quantifying method to determine the heating efficiency of the MPNs by measuring the rate of heat production in response to the magnetic field (Bahojb Noruzi et al., 2024; Pajoum et al., 2024).

Recent study has successfully designed a magnetic nano biocomposite scaffold through CMC-epichlorohydrin cross-linked interactions with the inclusion of silk fibroin (SF), HNTs and Fe_3O_2 MNPs which aim to make the magnetic nano biocomposite act as a hyperthermia cancerous cell eradication method (Eivazzadeh-Keihan et al., 2022). Furthermore, another study has also synthesized an acacia gum (AG)-SF hydrogel with the addition of polyvinyl alcohol and Fe_3O_2 MNPs to be implemented in the hyperthermia application as well (Pajoum et al., 2024). They have successfully proven the effectiveness of these nano-biomaterials in reducing and demolishing breast cancer line (BT549 cells) while maintaining the healthy human embryonic kidney cell line (HEK293T cells). Besides that, Bahojb Noruzi et al. (2024) has also fabricated a multifunctional nano biocomposite that combined the graphene oxide, casein, Zn-Al layered double hydroxide, sodium alginate, and Fe_3O_2 MNPs for the application of hyperthermia therapy, and reported the nontoxicity effect of the nano biocomposite, while maintaining its efficiency toward the human skin fibroblast cell (Hu02 cells) with an addition of an antibacterial properties.

Other than that, they have also shown the SAR value for each nano biocomposite in their paper, where Eivazzadeh-Keihan et al. (2022) and Bahojb Noruzi et al. (2024) reported the SAR value of 67 and 67.04 W/g, respectively, under a frequency of 400 kHz and 1 mg/mL concentration, while the SAR value of 93.08 W/g at 100 kHz and 1 mg/mL concentration was reported by Pajoum et al. (2024). These high values of SAR depict the efficiency of the modified nano biocomposite in its ability as a hyperthermia cancer therapeutic method, by increasing the temperature in the surrounding area in response to magnetic field frequency, depending on the application or the type of tumor on which they are applied. An elevated SAR value is achieved for deep-seated tumors, while superficial tumors prefer lower and higher frequencies of the magnetic field, respectively (Bahojb Noruzi et al., 2024).

11.6 Challenges and future perspectives

Despite the novel findings and unique characteristics of these smart biocomposite hydrogels in biomedical engineering field, there were still several issues and challenges that needed to be overcome in the process of developing these smart biocomposite materials. It has been noted that the difficulty in designing a artificial bone scaffold with high mechanical strength and flexibility, with consideration to the bone interface implementation behavior, remains a challenge in the tissue engineering field (Jia et al., 2023). This was due to the continual changes in the defect range of a bond defect area during surgery, which affects the integration of artificial bone scaffold (Hu, Yang, et al., 2024). Moreover, the lack of information regarding the biodegradability and long-term stability of these advanced biocomposites in the researched area needs to be acknowledged, since the breakdown by-products of these biocomposites could pose an issue to human health. Thus, these smart biocomposite hydrogels should have less biological effect on the implantation system for these materials to be applicable in the biomedical engineering sector. For instance, the biodegradability of a polymer was said to be the key factor in enabling a controlled and sustained release of the drugs, admitted to a targeted area, which is also the requirement for an eye's drug delivery system (Tsung et al., 2023).

Therefore an exploration of the nanoparticle utilizations (Namakka, Rahman, Bin Mohamad Said, et al., 2024; Namakka, Rahman, Mohamad Bin Said, et al., 2024) in improving the smart biocomposites' properties could be considered as a method in overcoming the problem, which simultaneously advances the current knowledge of smart biocomposite implementation in the biomedical engineering field. Magnetic nanoparticles have garnered the interest of recent studies due to their small size and higher surface area, excellent stability, easy surface modifications, high magnetic response ability, and low toxicity properties (Liu et al., 2020). Moreover, the combination of two stimulus-response for a smart biocomposite could also be a feasible approach in enhancing the current properties of smart biocomposites.

There has been an investigation on the dual-stimuli-responsive behavior of bionanomaterial through an anesthetic lidocaine loaded chitosan coated with nanoferrite ($CS/ZnFe_2O_4$), where the bionanomaterial was found to have pH and temperature responsive properties with excellent biocompatibility and sustained drug release features. However, this bionanomaterial could be further improved in terms of its degradability property and the probability of being widely studied, for it to be implemented in other fields in biomedical engineering applications, such as tissue engineering, with the cooperation of a multipurpose biocomposite. A combination of each of these material perspectives' unique characteristics could be a pathway for future advances in smart biocomposite materials.

Furthermore, the scalability of the smart biocomposite hydrogel is a topic that has been glossed over in the literature. This past research has not yet covered the topic of smart biocomposite synthesis process to industrial-scale practices or the ability of the smart biocomposite to transition from laboratory scale to industrial-scale production, which this review has not yet taken notice yet. Moreover, the issues on the cost-effectiveness of the synthesis process, the availability of the raw material, and the reproducibility of the smart biocomposite on a large scale should also be considered for a discussion in future studies. Hence, a detailed discussion on the economic and practical considerations of the smart biocomposites manufacturing is suggested to provide a higher valuable understanding of the application of smart biocomposite material in the biomedical engineering field.

11.7 Conclusion

This chapter has successfully discussed the application of smart biocomposites, which consist of a stimulus-responsive biomaterial in the biomedical engineering field. The application of pH-responsive, thermal-responsive, and magnetic-responsive biocomposites has been discussed in this chapter where the process of how these features helps in improving the implantation of scaffold in bone tissue engineering, a controlled wound dressing and drug delivery system, a precise anticancer drug delivery, and enhancing the hyperthermia cancer therapy was the highlighted insight of the smart biocomposites application in biomedical engineering field. Moreover, the issues faced in a smart biocomposites application have been pointed out, including an appropriate mechanical and flexibility properties of bone scaffold implantation for an irregular bone defect area, a lack of studies on the biodegradability breakdown of by-products for smart biocomposites, and the issue of industrial-scale production of these unique smart biocomposites. Therefore, a future pathway in which other researchers could implement has been stated in this chapter, where the utilization of nanoparticles with multipurpose biocomposites and stimuli-responsive biomaterials could be explored further to obtain a new and novel material that could further improve the current technology of smart biocomposites. Additionally, dual-stimulus-responsive biocomposites, a combination of two different stimulus-responsive biomaterials, could also be investigated as a novel approach in the smart biocomposite materials for the biomedical engineering field.

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