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Gelatin-Based Biocomposites for Skin Regeneration and Wound Repair

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ABSTRACT

Chronic skin injuries constitute a serious medical problem, since they suffer from prolonged inflammatory response, angiogenesis impairment, bacterial infection, and delayed regeneration of tissues. Traditional skin dressings usually offer only passive protection and cannot regulate the local microenvironment actively. Biomaterial-based approaches, in contrast, can serve as efficient methods of skin regeneration. Among the natural biomaterials, gelatin attracts much attention due to good biocompatibility, biodegradability, the ability to mimic the extracellular matrix (ECM) and the ease of manufacturing into various forms, such as hydrogels, scaffolds, nanofibers, and bioinks. However, the low mechanical stability, fast degradation rate, and weak antibacterial effect of gelatin do not allow using this substance directly in medicine. That is why modern research concentrates on gelatin biocomposites, which consist of gelatin and various natural or artificial polymers, inorganic nanoparticles, and bioactive components. This article gives an overview of the structural features and biological properties of gelatin, describes recent advances in the application of gelatin biocomposites for skin regeneration and wound healing, and explains their modes of action and antimicrobial effects. Furthermore, current challenges related to large-scale production, standardization, controlled drug delivery, and clinical translation are discussed, together with emerging strategies involving smart hydrogels, multifunctional nanocomposites, and 3D bioprinting technologies.

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1. Introduction

Acute and chronic wounds continue to pose a significant problem in global health care due to the prevalence, long-term healing periods, and socio-economic costs [1-3]. Whereas an acute injury goes through the typical stages of healing, the chronic injury typically does not advance through the healing process and is often stuck in the inflammatory stage [4, 5]. The high rates of diabetes, obesity, vascular diseases, and aging have contributed to the rise in chronic wounds. Thus, it is necessary to improve the treatment methods [6-8]. Wound healing is a dynamic and highly coordinated biological process involving four overlapping phases: hemostasis, inflammation, proliferation, and remodeling [8-11]. Effective healing necessitates a coordinated action of inflammatory cells, fibroblasts, keratinocytes, endothelial cells, extracellular matrix (ECM) proteins, and various cytokines. But inflammation, infection by bacteria, oxidative stress, and defective angiogenesis often interfere with this process, causing delayed wound healing and chronic wounds [11-15]. Wound dressings made from materials like gauze and cotton merely serve the purpose of offering protection and do not have much of a role to play in regulation of the wound microenvironment or regeneration of tissues. This is why there has been so much focus on biomaterials-based wound dressings, as they offer all these functions and more [13, 16]. The use of hydrogels, electrospun nanofibers, nanoparticles, and bioactive scaffolds has been found to be promising in improving wound healing results due to their enhanced antimicrobial activity, controlled drug delivery, and tissue regenerative ability [17-19].

Among several materials being researched for skin regeneration, gelatin has turned out to be among the most promising of natural polymers due to its superior properties such as biocompatibility, biodegradability, non-immunogenicity, low cost and structural analogy with natural extracellular matrix [20]. Gelatin is a denatured form of collagen, which has many cell binding sequences that help in cell adhesion, proliferation, and migration. Due to these properties, gelatin is very useful for wound healing and tissue engineering [21, 22]. The clinical use of pure gelatin, on the other hand, is limited due to its poor mechanical strength, high degradation rate, poor structural stability, and lack of antibacterial activity [23, 24]. To overcome these shortcomings, many functional materials, such as natural and synthetic polymers, bioactive ceramics, metal and metal oxide nanoparticles, carbon-based nanomaterials, therapeutic agents, and growth factors have been used along with gelatin to form multifunctional biocomposites based on gelatin [25-27]. In addition to providing biological activity, such as antibacterial, antioxidant, anti-inflammatory, angiogenic, and controlled drug delivery properties, these systems contribute to enhanced mechanical strength and degradation performance of gelatin [28-31]. Because of these benefits, the bio-composite materials based on gelatin have been recognized as excellent candidates for the fabrication of novel wound dressing materials that can effectively control the wound microenvironment [30, 32]. Even though many review papers have been published concerning gelatin-based biomaterials for wound repair, these papers have been mainly concerned with particular formulations, production techniques, or potential applications of these biomaterials. It should be noted that until now no up-to-date and comprehensive review paper that gives an overview of the biological behavior and action modes of gelatin-based biocomposites composed of natural and synthetic polymers has been written.

In this review, recent advances in gelatin-based biocomposites for skin regeneration and wound repair are summarized, covering

gelatin's properties, limitations, fabrication methods, and biological performance. Multifunctional composites with enhanced therapeutic activities are highlighted. The clinical obstacles and prospective pathways are also thoroughly examined.

2. Gelatin as a Biomaterial for Skin Tissue Engineering

Gelatin has been among the most investigated natural biomaterials in skin tissue engineering research over the past few years owing to its exceptional adaptability and diverse use in regenerative medicine [30, 33]. Gelatin has now been able to be used in many tissue-engineering products such as 3D printing, bioactive composite scaffolds, and photocrosslinkable hydrogels, making it an ideal candidate for creating biomaterials with improved biological properties [34-36].

Gelatin is a natural polymer which is made by partial hydrolysis of collagen, a protein that exists in high amounts within mammals. The process of partial hydrolysis of collagen breaks the highly ordered triple helix structure of collagen leading to formation of polypeptides having unique physicochemical properties. The type of gelatin depends on the method of extraction where gelatin may be either type A or type B, depending on the isoelectric point and physicochemical properties [37]. Due to its unique structure, gelatin is easily transformable into different forms such as hydrogels, porous scaffolds, films, microspheres, and bioinks, which makes it a very flexible biomaterial in tissue engineering and regenerative medicine applications [35, 38].

The extensive use of gelatin is mainly due to its superior biological performance. Being a biomaterial derived from collagen, gelatin displays excellent biocompatibility, very low immunogenicity, and remarkable biodegradability, which makes it very applicable in tissue engineering [37, 39]. It is possible to break down via matrix metalloproteinases enzymes, thereby making scaffold breakdown happen in conjunction with tissue formation [40]. Additionally, it has cell binding sites due to the presence of Arg-Gly-Asp (RGD) sequences. In addition to having similarities with the ECM, this characteristic also creates an ideal environment for cell processes such as adhesion, proliferation, and migration [41].

Nonetheless, the clinical use of gelatin alone faces several constraints due to a series of intrinsic disadvantages that come along with it. Firstly, gelatin has comparatively weak mechanical properties and instability, which makes it impossible to be used directly in skin tissue engineering [42]. Secondly, gelatin degrades too fast, has high water solubility and swelling ability, and therefore can disintegrate before the completion of tissue engineering [39, 43]. Also, gelatin does not have antibacterial properties and thermal stability. Thus, the chemical modification of gelatin and combination with natural or artificial polymers, inorganic bioactive compounds, nanoparticles, and other functional components are widely used [44, 45].

3. Gelatin-Based Biocomposites

The use of gelatin as a natural biomaterial for wound dressing applications is one of the areas where extensive research has been carried out due to its superior biocompatibility, biodegradability, low immunogenicity, and cell adhesion and proliferation property. Nevertheless, although these are some of the beneficial properties exhibited by gelatin, its use in wound dressing applications has been constrained by several disadvantages, such as weak mechanical strength, enzymatic degradation, poor stability in

physiological environment, and moisture deformation [39, 46, 47]. To address these issues, gelatin biocomposites have been created using gelatin with other biomaterials, which include natural and synthetic polymers, inorganic nanoparticles, and bioactive components. By using such composite structures, one can take advantage of the biological properties of gelatin and add new

features like enhanced mechanical strength, degradation rate, antibacterial properties, antioxidative properties, and regeneration capacity. Hence, the use of gelatin biocomposites is highly recommended for wound dressing materials in chronic wounds, especially there, where inflammatory response and tissue reconstruction need to be managed simultaneously (Fig. 1) [48].

3.1. Gelatin/Natural Polymer Biocomposites

Natural polymers are one of the most widely used materials that are mixed with gelatin due to their high biocompatibility and structural similarity to the native extracellular matrix (ECM). Gelatin mixture with natural polymers like chitosan, alginate, hyaluronic acid, collagen, and silk fibroin increases scaffold stability, water content, cell interactions, and biological function. Such mixtures produce biomimicking matrices that create an environment that supports proliferation of fibroblasts, migration of keratinocytes, formation of new blood vessels, and remodeling of the ECM [39, 49, 50]. This compound is often used in gelatin systems because of its antibacterial, hemostatic, and immunomodulation properties. The gelatin-chitosan composite materials have been shown to perform better mechanically and have been proven effective at protecting against bacterial infection, thus being suitable for treating infections in wounds. The positive charge of the amino groups in chitosan allows it to interact with the negative charge of cells [48].

Alginate is yet another popularly used biopolymer in wound dressings made from gelatin due to its high water-absorbing capacity and its ability to keep the wound environment moisturized. The composite of gelatin and alginate will be able to absorb any excess fluid from the wound while at the same time retaining enough moisture for cell proliferation and development of granulation tissue. Moreover, alginate-based systems can control the release of loaded drugs [51].

Hyaluronic acid, an essential component of the native ECM, has also been integrated with gelatin to enhance regenerative responses. The combination of gelatin and hyaluronic acid promotes cellular migration, angiogenesis, and collagen deposition by providing biochemical signals similar to those found in natural skin tissue. Similarly, collagen and silk fibroin-based gelatin composites have shown improved cell adhesion and mechanical properties due to their structural similarity to dermal proteins, supporting their application in skin tissue engineering and wound repair [48]. Overall, gelatin/polysaccharide-based composites offer biomimicking technology for wound treatment using a combination of the biological properties of gelatin along with the additional properties of polysaccharides. The systems ensure the creation of a beneficial environment for tissue regeneration while retaining high biocompatibility.

3.2. Gelatin/Synthetic Polymer Biocomposites

Despite having great biological properties, the low mechanical properties and quick degradability of gelatin limit its use in scenarios that require structural durability. Hence, synthetic polymers have been introduced to gelatin matrices to strengthen the mechanical and degradability properties while retaining the biological properties of the gelatin [46, 52, 53]. Moreover, the integration of synthetic polymers, such as PCL and PLA, provides even more strength and degradation properties to the gelatin scaffolds to be applied in the wounds for long periods of time [52-54].

The combination of Gelatin/PCL has been shown to have several advantageous features for the purpose of skin regeneration because of its ECM-like structure, high surface area, and the capability of supporting cell growth. While PCL enhances the mechanical strength and provides controlled degradation rate, Gelatin offers cell recognition motifs and higher hydrophilicity [55]. To address the limitations associated with gelatin, several composites have been produced using the combination of gelatin with different natural polymers, synthetic polymers, nanoparticles, and bioactive materials. The gelatin composites have shown increased mechanical strength, improved biological activity, and multifunctionality for skin regeneration and wound healing. Some examples of the gelatin composites reported recently are listed in Table 1.

4. Mechanisms of Skin Regeneration

The biological and physicochemical effects of gelatin biocomposites lead to skin regeneration, such as regulation of inflammation, promotion of angiogenesis, stimulation of cellular activities, and reconstruction of extracellular matrix. Current research shows that functionalized GelMA biocomposites could regulate the wound environment through provision of a supportive matrix for cell migration and interactions of immune cells with regenerative tissues [56, 57].

The inflammatory phase is characterized by an increase in the activity of macrophages and oxidative stress which may inhibit proper wound healing. Recently developed advanced hydrogels based on gelatin have been designed to control macrophage polarization and minimize inflammation.

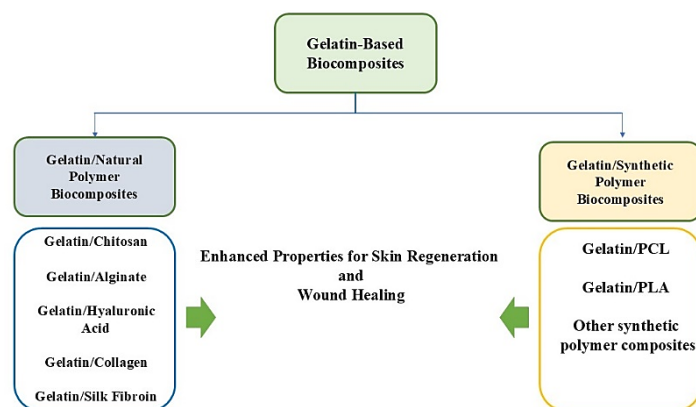


Fig. 1. Classification of gelatin-based biocomposites for skin regeneration and wound healing.

Table 1

Recent advances in gelatin-based biocomposite systems for wound healing and skin tissue regeneration.

Main composition	Bioactive Agent	Swelling Capacity	Pore Size	Function	Ref
Pectin methacrylate (PeMA) / GelMA hydrogel	Curcumin	~90%	43 μm	pH-responsive drug release, antibacterial activity, wound healingpH-responsive drug release, antibacterial activity, wound healing	[58]
Gelatin/Chitosan/Hyaluronic acid	Buddleja globosa extract	~2500%	70–154 μm (average ~111 μm)	Antimicrobial activity and fibroblast biocompatibility	[59]
Gelatin/Chitosan/Cellulose nanocrystals	Cellulose nanocrystals	~1071%	~148 μm	Supports human dermal fibroblasts and keratinocytes for wound healing	[60]
Collagen/Hyaluronic acid/Gelatin	None	>20 g water/g dry scaffold	132.5 $\pm$ 8.4 μm	Skin regeneration and support of human skin cells	[61]
Carboxymethyl chitosan/Gelatin bilayer scaffold + PLLA nanofibers	Protocatechuic aldehyde–Ga ³⁺ (PA@Ga)	2000%	78–138 μm	Hemostasis, antibacterial activity, wound healing, skin regeneration	[62]
Chitosan/Gelatin/Hyaluronic acid hybrid scaffold	Graphene oxide + Silymarin	High	118.5 μm	Full-thickness burn wound repair, antibacterial, antioxidant, tissue regeneration	[63]
GelMA-NBNAGA/LAP hydrogel	Rifampicin	58.6 $\pm$ 1.9 g/g	4.3 $\pm$ 1.4 μm	Antibacterial activity and infected wound healing	[64]
Gelatin/Chitosan scaffold	Aloe vera + Snail mucus	highest	93–29 μm	Skin tissue engineering; wound healing; enhanced	[65]

One such example is a GelMA hydrogel containing macrophage-derived exosomes that enhance immune modulation and angiogenesis [56, 66]. Angiogenesis is a vital component in gelatin wound repair since the new blood vessels that develop provide oxygen and nutrients necessary for tissue repair. Recent GelMA-based scaffolds have proven to increase the activities of the endothelial cells as well as angiogenesis due to their biomimetic composition and delivery of bioactive components [57, 67]. The re-epithelialization process is another key pathway which is facilitated through the application of gelatin-based materials. GelMA bandages facilitate keratinocyte attachment and migration to enhance barrier function recovery. Furthermore, the use of antioxidants and antimicrobial drugs within gelatin matrices helps decrease oxidative stress and prevent infections respectively [68, 69]. Overall, it has been shown through recent developments that not only can biocomposites based on gelatin be used as a passive dressing of wounds, but they can also be employed as an active system that regulates the immune response and provides a suitable microenvironment for skin regeneration. Rahimi et al. [70] created taurine-conjugated gelatin hydrogel (Gel-Ph-Tau), which played a major role in skin regeneration by inducing endothelial cell migration and angiogenesis. The presence of taurine conjugation enhanced the mechanical properties and degradation of the hydrogel, increased the expression of proangiogenic genes such as VEGF, bFGF, and Ang-1 in addition to the endothelial marker genes CD31 and CD44. A study conducted by Jaber et al. involved the development of granular hydrogel scaffolds based on gelatin methacryloyl (GelMA) for skin wound healing purposes. This scaffold had the advantage of having interconnected macropores and a conducive microenvironment for tissue regeneration, with increased wound closure observed in vivo [57]. Kim et al. [71] used electrospinning technology to synthesize gelatin/polyurethane nanofibers for wound dressing applications. The nanofiber membranes demonstrated acceptable mechanical strength, water uptake, and outstanding cell performance that facilitated fibroblast and keratinocyte attachment and proliferation.

5. Current Challenges and Future Perspectives

However, despite the tremendous advances that have been made in the synthesis and use of gelatin biocomposites for wound

healing applications, there exist some issues which have made the transition of these biocomposites to clinic rather difficult. The issues include poor mechanical strength, batch-to-batch variations, high rate of biodegradation, and the inability to control the release of drugs [72, 73]. Current developments suggest that future studies should be aimed at exploring the design of multi-functional gelatin biomaterials which can respond to the change in pH, reactive oxygen species (ROS), temperature, or enzymes in the wound microenvironment. These smart dressings will be able to deliver drugs on demand while simultaneously controlling and modulating the healing process [74, 75]. A more encouraging approach involves the use of GelMA bioinks for the fabrication of personalized skin constructs with well-controlled designs using 3D bioprinting technology. However, there are several problems that need to be solved to translate this technology into clinic applications. These issues include, but are not limited to, bioink development, vascularization of large tissue structures, scalability, quality control, and regulatory considerations [76, 77]. Overall, the combination of the above-mentioned factors is likely to facilitate the creation of the next generation of gelatin-based biocomposites, which would improve the treatment of acute and chronic skin injuries.

6. Conclusion

Biocomposites based on gelatin have become one of the most promising biomaterials used for skin regeneration and wound healing because of high biocompatibility, biodegradability, and ECM-mimetic properties. Gelatin/chitosan/hyaluronic acid scaffold loaded with Buddleja globosa extract offers a strong combination of high swelling capacity, antimicrobial/biological activity, fibroblast support, and skin tissue regeneration, making it well suited (in our study, the best option) for wound healing. The combination of gelatin for cell adhesion, chitosan for antimicrobial/hemostatic effects, and hyaluronic acid for hydration and tissue repair makes it a well-balanced scaffold. Despite the presence of some disadvantages of natural gelatin like weak mechanical strength and fast degradation, these can be successfully resolved using the combination of natural or artificial polymers, nanoparticles, and bioactive compounds. Multifunctional biocomposite systems will not only allow improving physical and

chemical properties of gelatin-based materials but also actively control important biological processes associated with wound healing such as inflammation, angiogenesis, matrix remodeling, re-epithelialization, and antimicrobial protection.

The development of smart hydrogels, nanocomposite dressings, and 3D bioprinting technologies is another factor contributing to the future success of gelatin-based biomaterials and approaching them to clinical application. However, there are still several unresolved issues associated with large-scale production, storage, standardization, and regulatory approval of these materials before they could be widely used in practice. Further interdisciplinary research combining biomaterials, bioengineering, nanotechnologies, and regenerative medicine will contribute to the creation of new-generation gelatin-based biocomposites and wound therapy methods.

Author contributions

Darya Nejadkoorki was responsible for conceptualization, writing the original draft, and writing, reviewing, and editing the manuscript. **Tengiz Tkebuchava** contributed to writing the original draft and reviewing and editing the manuscript. **Parisa Yadollahi** contributed to writing, reviewing, and editing the manuscript and supervised the study.

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Conflict of interest

The authors declare that they have no conflicts of interest to disclose.

Data availability

No datasets were generated or analyzed during the present study; therefore, data availability is not applicable.

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