

Syntheses and Fabrication of Sensitive Bioactive Surgical Sutures from Human Hair Based on polyvinyl Alcohol/graphene Oxide Composite for Biomedical Applications

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RECENTLY material science has become eye-catching to scientists, engineers, and researchers as a modern technology. Surgical sutures since ancient times have been the best solution for treating wounds. The risks associated with and limitations of wound closure devices demanded the need for cost-effective techniques and efficiency for wound healing. There has been a large-scale evolution and expansion of the material research and business around biomedical applications. Until now, sutures and staples have been the main used tools in the biomedical industry. Absorbable sutures which are based on biopolymer more preferred than nonabsorbable ones. Absorbable sutures

(AS) are degraded within the body usually by the aid of proteolytic enzymes or with hydrolysis. Although they are biocompatible, there are limitations in the lack of sufficient antimicrobials, and drug delivery which are desired for biomedical applications. This study aimed to modify the absorbable polyvinyl alcohol and chitosan as biopolymer mixed with GO as a nanomaterial and keratin as a source of protein (PVA /Cs/GO/ Ke) sutures to support an antimicrobial effect that is protected from multidrug-resistant microorganisms. In addition, graphene oxide has promised material for suture enhancement. This could be because GO provides active chemical sites. GO has also been used to coat surgical sutures to improve their functionalities to prevent bacterial adherence. So, it's a good antimicrobial activity by itself or its drug delivery. Chitosan is a qualified antimicrobial agent can be attributed to its cationic nature and reduced water content. The mixture was turned to nanofiber by an electrospinning process. Then the fiber was turned to multifilament by a twisting machine. The results revealed our main advantages the first, absorbed nanofiber sutures with strong antimicrobial against pathogenic strains the second, absorbed time controlled the third cheapest biomaterial and last but not least AS is ecofriendly which depends on human hair.

High biocompatibility of biopolymers,
Economic cost for the surgeon,
Self-sterilization without compromising material integrity,
Absorption with bioactive sites,
AS has bacterial resistance.

Keywords: *Sutures, graphene Oxide, PVA, Nanotechnology, antimicrobial.*

Introduction

Sutures play an important role in facilitating wound healing and in ensuring surgical interventions succeed. Suture-associated surgical site infection could be done when pathogens grow onto the suture surface and make resistant

biofilms to antibiotic treatment. Thus, the rates of morbidity will increase and are accompanied by high mortality[1].

Microbial growth onto the sutures is based on the microbial species due to infection and the chemical structure of the suture's material [2–5].

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Multifilament and monofilament sutures have been reported. Although multifilament is stronger than monofilament [6], some studies indicate that multifilament sutures are more exposed to bacterial adherence than monofilament, which can cause severe inflammations [7, 8]. So, the sutures must be strong with no bacteria growth.

Suture natural polymers such as collagen, silk, fibrin, carboxyl methyl cellulose (CMC), hyaluronic acid (HA), and chitosan which have good activities with cells through cell surface receptors [9] but they are expensive, have lower mechanical properties, and not easily accessible [12].

The US Food and Drug Administration (FDA) confirmed the use of a polyvinyl alcohol (PVA) implant for surgical application [10] and approved that is safe and biocompatible [11]. So, PVA has been used as a backbone in many suture techniques [12, 13]. Although poor mechanical properties of PVA have been improved by chitosan, it still exhibits poor mechanical properties [14].

Nanomaterials could introduce many solutions for problems in many fields. So, in sutures, nanoparticles supported both physical and biological properties. Suture materials treated with antimicrobial agents are an important approach for keeping wounds safe from infections. Most antibiotic drugs come with risks to public health and associated mortality [15–18]. It comes from antibiotic side effects or leads to the development of bacterial resistance [19–20]. So new materials must be developed to avoid antimicrobial resistance.

Many papers have been published for enhancing surgical suture materials such as non-absorbable silk sutures coated with silver nanoparticles (NP) as an antimicrobial agent [21–26], zinc oxide NP impregnation on surgical sutures to support wound healing [27], gold nanoparticles slurry was dipping on surgical suture to improve the antibacterial effects [28].

Over the past decade, graphene oxide has been the most investigated for its amazing properties. Many studies prove that graphene-oxide has a high surface area [29–32], many functional groups [33–35], protein adsorption [36–39], antimicrobial potentials [40–45], hydrophilicity properties [34], [44, 45] and flexible to handle, so they were used for wound dressing to prevent infection in different wounds [41], [46–52]. When addition small quantity of GO (< 5%) enhanced

the mechanical properties of the wound dressing and protein affinity which wound fast healing, and support of bone tissue generation.

Advanced green technology methods that aim to treat the waste are known to be environmentally, safe nontoxic, cheap, and practical applications with appreciable results for industry [53].

Human hair is one of the interesting biological fibers [54–56]. Many tons of human hair are wasted in the world annually; so, they lead to an environmental challenge [57]. Waste human hair trade has increased day after day. The economics of the human hair industry reached \$7 billion in 2020 as a commercial application of wasted human hair fiber [58]. The basic backbone of hair is incredibly strong keratin; a single strand of hair can load 100-150 grams of weight [59, 60]. In addition, the elasticity of the hair makes it able to restore its original position after removing the deformation load. This leads to the fact that all human hair kinds can support reinforcement [60]. Protein is found in the greatest quantity in hair with low Sulphur content. Keratin is a protein found in the cortex. Keratin has 18 amino acids such as cysteine, glutamic acid, serine, glycine, threonine, leucine, valine, arginine, and isoleucine. In other terms sutures/Keratin is fabricated by the hot-melt extrusion method at 63 ± 1 °C [61,62]. On the other hand, increasing keratin temperature leads to the deformation of keratin.

Materials and methods

Materials

Hydrochloric acid 37% GR for analysis ACS (IL, Naser, analytical purity) and sodium hydroxide powder (IL, Sigma, analytical purity) were employed to hydrolyze wool fibers. Chitosan (Cs) and Polyvinyl alcohol (PVA) powder, acetic acid.

Methods

Preparation of absorbable sutures (AS) Cs/PVA/GO

Cs (1gm) was dissolved in 30 ml of acetic acid solution with agitation until the solution *turned clear. *chitosan reduces moisture and water uptake [63]. Then aqueous solution of PVA was prepared by dissolving 10gm of PVA in 50 ml of water. The mixture was prepared with 1 ml of Cs and 50 ml of PVA by stirring for 1 hour at 60 C temperature to reach a clear mixture.

GO was prepared by Hummer method modification. Graphite (2.0g) was added to sulfuric acid concentrated H₂SO₄ (50 ml) under stirring in an ice bath, followed by the addition of

NaNO₃ (1 g), and then the mixture was cooled to 1 °C. Finely KMnO₄ (6.0 g) was added to the solution for 2 hours then the solution agitates for 4 hours at room temperature.

Preparation of Keratin (Ke) from human hair

The hair was prepared by washing with DW and ethanol. The clean hair was immersed in Alkali and acid solution to determine which solution was more active in extraction of keratin than the other. The keratin protein was indicated by SEM image.

The influence of the temperature on alkali solution for keratin extraction

The alkali solution at Ph. 12 was adjusted by caustic soda. One gram of hair was dissolved in sodium hydroxide solution after washing many times with DW. Then raise the temperature at 80°C to rapid the hydrolysis of the hair for 5h. In the second solution, one gram of hair was immersed in sodium hydroxide solution at Ph 12 for 24h for complete hydrolysis at room temperature.

The influence of the temperature on the acid solution for keratin extraction

The acid solution at Ph* 1 which Ph was adjusted by hydrochloric acid. One gram of hair was dissolved in 50 ml dilute hydrochloric acid solution by raising the temperature to 80°C to rapid the hydrolysis of the hair for 5h. In the second

step 1 Cs (1gm) was dissolved in 30 ml of acetic acid solution with agitation until the solution.

step 2 GO was prepared by Hummer method modification.

solution, one gram of hair was immersed in cold hydraulic acid solution at Ph 2 for 24h.

PVA /Cs /GO/ Ke solution preparation

1 ml of GO (1%) was added to PVA /Cs for one hour. Then, 1ml of Ke was added to the mixture PVA /Cs /GO under agitation at 60 °C.

Electrospinning of PVA /Cs /GO/ Ke.

We used 1 gm of the boy's hair of brown color.

In the electrospinning technique, a PVA /Cs /GO/ Ke solution is driven through a dispensing needle that is in front of aluminum foil which covers a cylindrical metallic fibers collector, then an electric field is applied between the needle and aluminum foil by the application of a potential difference. PVA /Cs /GO/ Ke fibers were loaded to manufacture bioactive sutures, which had a greater affinity for tissues while providing antimicrobial protection. Cs (1gm) dissolved in 30 ml of acetic acid solution with agitation and Hummer method modification is applied to prepare GO. Figure 1 Indicates an XRD of GO and a TEM image of the prepared GO.

Figure 2 shows the electrospinning system. The applied potential of the electrospinning system was 20 KV, the rate was 4ml/h, and the distance between the needle and the collector was 30cm.

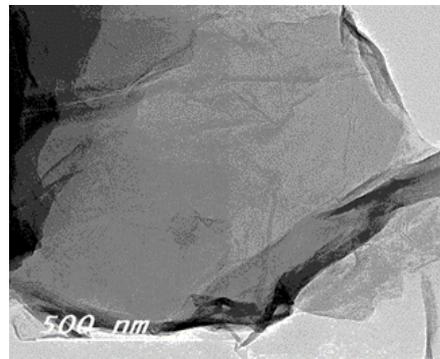
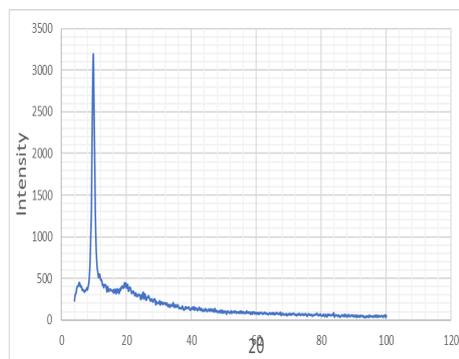


Fig. 1: a) XRD of GO, and b) TEM image of the prepared of GO.

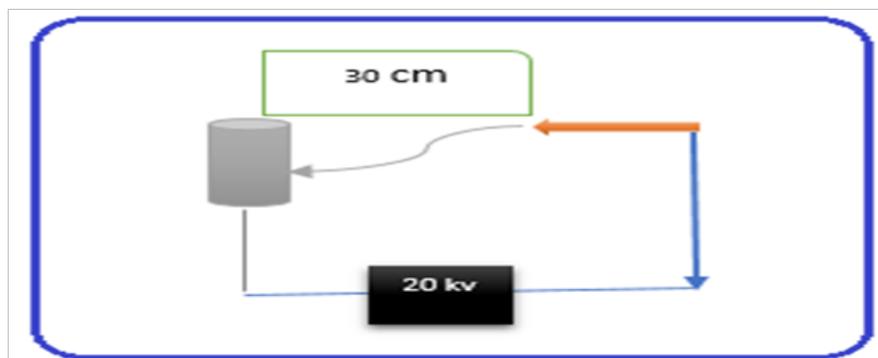


Fig. 2. The electrospinning system.

Characterization of PVA /Cs /GO/ Ke

ESM (Jeol) fine layer coating for *prepared the sample with a thin layer of gold, Scanning electron microscopy (SEM) (Jeol), and Energy Dispersive X-ray (EDX). Center lab, University of Mania, Mania, Egypt while the facility of

Degradation study

The degradation of AS PVA /Cs /GO/ Ke was assessed by the phosphate buffer solution (PBS) as reported earlier [20]. The AS nanofiber suture was exposed to PBS to indicate the biological stability period of the sample and its rate of degradation. The level of degradation was estimated for the nanofiber suture sample *depending on weight loss.

Morphological and microanalytical characterization

To analyze the elements and structure, the SEM captured images of the PVA/Cs/GO/Ke nanofiber sutures. These were affixed onto the SEM holder using carbon tape. To obtain clear images and EDS peaks for elemental analysis, the sutures were not coated with a thin layer of gold for conductivity. The SEM utilized a JSM IT200 Field Emission Scanning Electron Microscope (JEOL, Japan) with an accelerating voltage ranging from 0.3 to 30kV, depending on the necessary magnification, to capture these images.

step 3 10 gm of PVA in 50 ml of water.

Step 4 Preparation of Keratin (Ke) from human hair by different methods .

Antimicrobial activity

The antibacterial and antifungal properties of PVA/Cs/GO/Ke nanofiber sutures were tested against different microorganisms including Escherichia coli (a type of gram-negative bacteria), Candida albicans (a fungus), and Staphylococcus aureus (a type of gram-positive bacteria). To experiment, C. albicans was cultured in Saboraud Dextrose Broth (SDB), while E. coli and S. aureus were cultured in Nutrient Broth (NB)

Results and Discussion

The findings presented in the Figures showcase the outcomes obtained from Energy Dispersive X-ray Spectroscopy (EDS) and Scanning Electron Microscopy (SEM) imaging. The SEM images of the PVA/Cs/GO/Ke nanofiber unveiled the characteristic multifilament arrangement of the sutures, highlighting the deposition of GO nanoparticles onto the biopolymer. Additionally, the presence of Ke, a protein within the sutures, was identified, along with the detection of nitrogen ions. we added 10gm of PVA in 50 ml of water to prepare a Keratin (Ke) from human hair by different methods as shown in Figure 3.

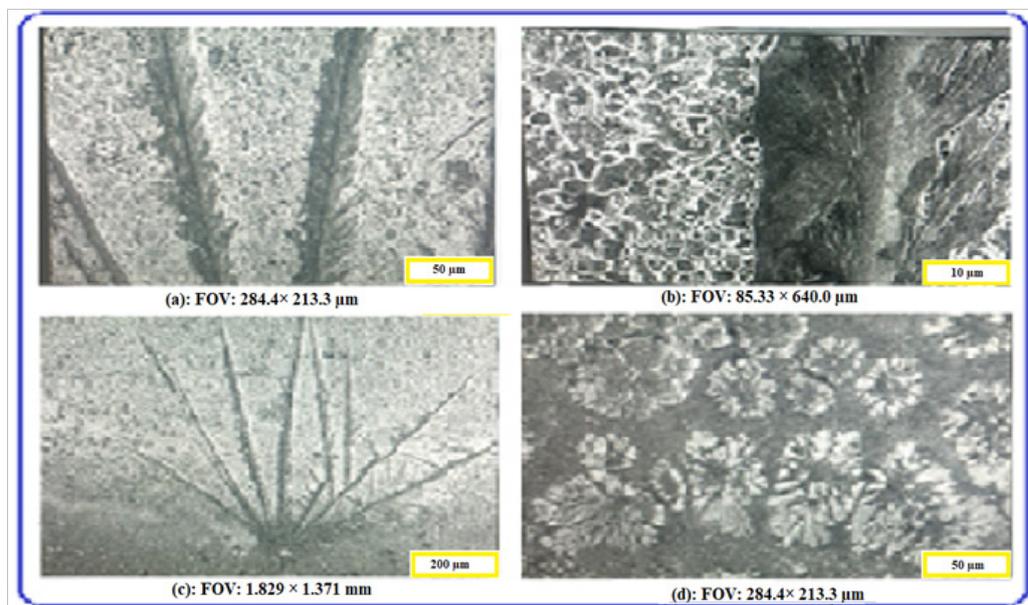


Fig. 3. The hydrolysis of hair: (a) under alkali solution (80°C), (b- c) hair hydrolysis within 30 min, and (d) hair hydrolysis after 5h.

Figure 3 a,b and c show of hydrolysis the ke formation at different scales in the start of hydrolysis . Alpha keratins (hair keratin) are fibrous structure, and looks like the groups of thread of a screw, after complete hydrolysis this structure disappeared as shown in figure 3d which shows the use of NaOH at high temperatures the Ke was damaged after completed hair hydrolysis. While the use of NaOH at room temperature kept Ke safe from damage as shown in Figure 4.

Figure 5 shows the influences of HCl for Ke hydrolysis Figure 5 -a indicates the human hair didn't hydrolyze in HCl at room temperature after 24 h **Figure 5-** b,c indicates the influences of HCl for Ke hydrolysis at high temperature which the human hair hydrolyzes in HCl at 80 °C after 5h. *biodegradation*

n fact, the degradation experiment, and physiological case were done with phosphate buffer solutions at pH 7.4 for 6 weeks. It indicates that the degradation of the suture supports the diffusion of the GO and, therefore, inhibits the bacterial and super bacterial which have protection against antibiotics. PVA /Cs /GO/ Ke nanofibers absorbed sutures have soft surfaces and antimicrobial structures supporting the results in the conclusion confirming that sutures are suitable for a wide range of types of wounds. The rate of degradation indicates that the suture lost 61% after two weeks from its weight, 74% after 3 weeks, and complete degradation was observed after 5 weeks.

Nanofibers

Figure 6 shows AS nanofibers under the electrospinning process. PVA /Cs /GO/ Ke nanofibers images were done by scanning electron microscopy. These images indicate multifilament nanofiber sutures.

Figure 7 shows AS nanofibers under the electrospinning process. PVA/Cs/GO/ Ke nanofibers were compared with PAN nanofibers. PVA /Cs /GO/ Ke nanofibers gave flexible straight strain with good mechanical properties. On the other hand, PAN nanofibers gave short stains and random formation with weak mechanical properties.

Conclusion and future

perspectives

Conventional suture materials commonly have drawbacks such as their inclination to develop microbial biofilms, leading to increased health complications. Recent research has concentrated on bioactive sutures, which promote tissue regrowth and possess antimicrobial characteristics. Suture materials play a vital role in the field of biomedicine, with factors like ease of use, cost-effectiveness, compatibility with the body, antimicrobial traits, and mechanical strength significantly influencing their overall quality. Absorbable sutures like AS Cs/PVA/GO/Ke offer additional benefits as they are renewable, biodegradable, environmentally friendly, biocompatible, and exhibit reduced antigenicity.

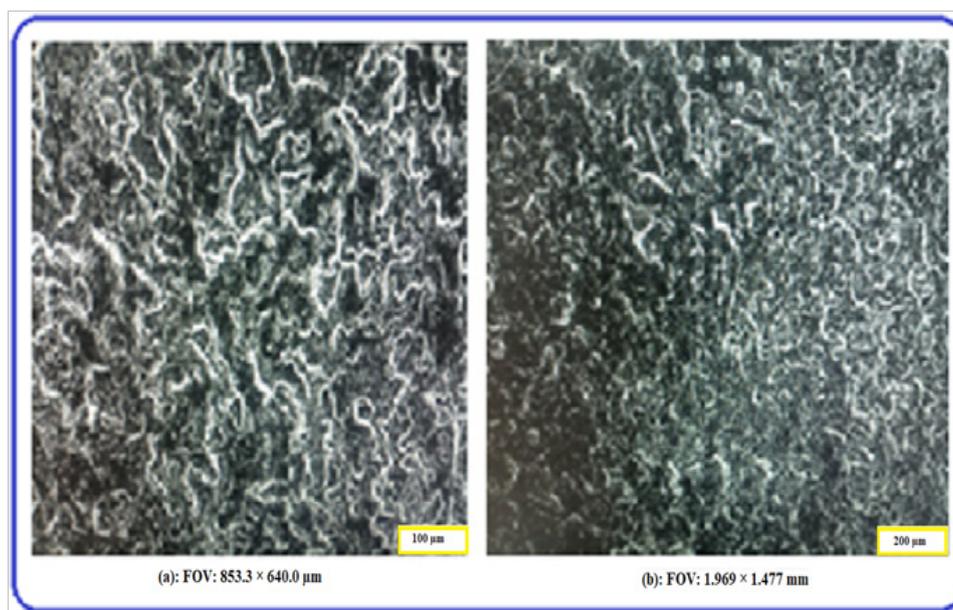


Fig. 4. The hair hydrolysis in NaOH at 25°C after 24h at different magnifications(a,b).

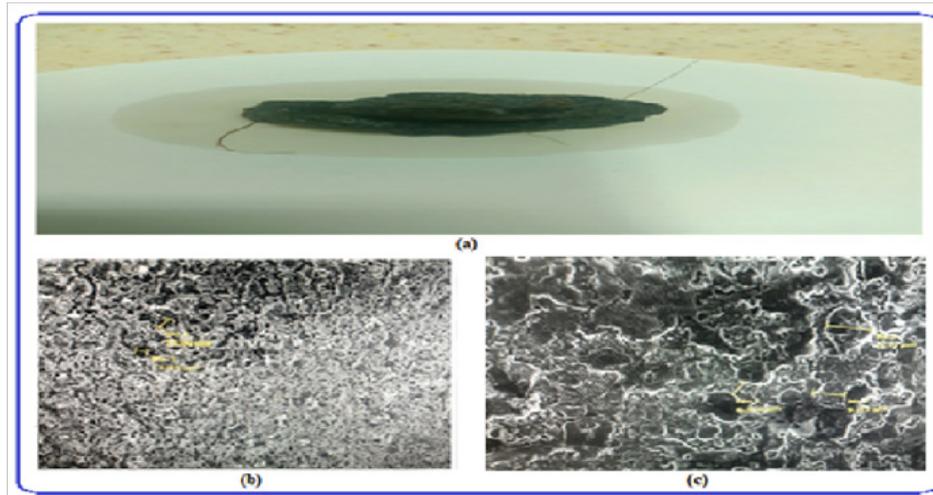


Fig. 5. The washed hair in HCl then hydrolysis in NaOH at 25oC after 5 min at different magnifications from (a-c).

Step 5 Physical mixing PVA /Cs /GO/ Ke solution preparation.

Step 6 PVA /Cs /GO/ Ke nanofibers preparation by electrospinning technique.

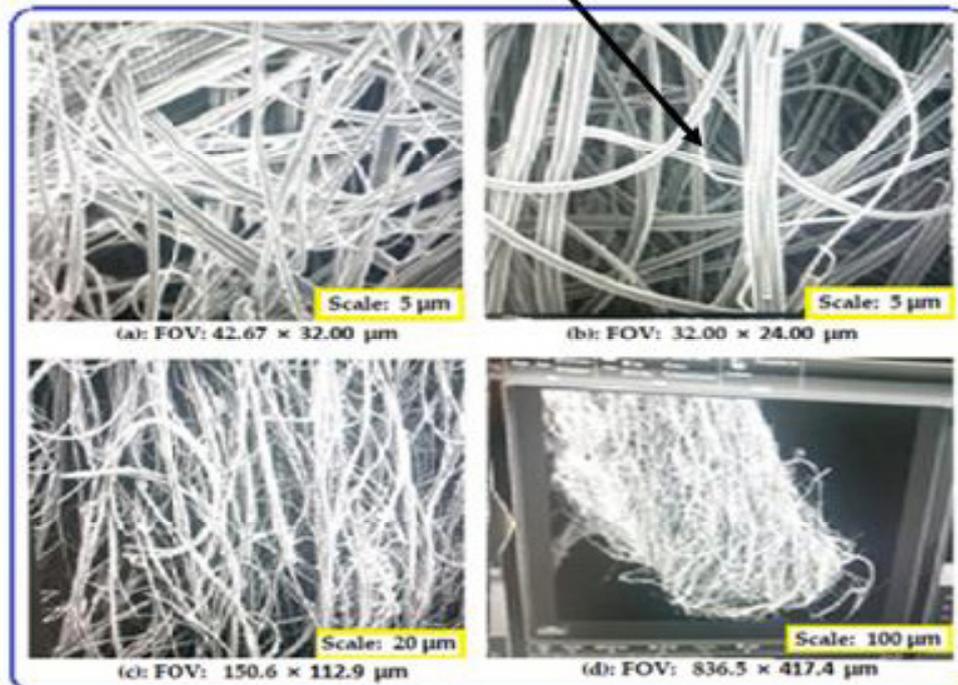


Fig. 6. The nanofiber sutures at different magnifications from (a-d).



Fig. 7. AS nanofibers under the electrospinning process(a,b).

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تخليق وتصنيع الخيوط الجراحية الحساسة النشطة بيولوجياً من الشعر البشري استناداً إلى مركب GO/PVA للتطبيقات الطبية الحيوية

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في الآونة الأخيرة أصبح علم المواد من التكنولوجيا الحديثة التي تلفت أنظار العلماء والمهندسين والباحثين. وفي المجال الطبي والجراحة فإن الغرز الجراحية هي الحل الأمثل لعلاج الجروح منذ القدم. وتتطلب المخاطر المرتبطة بأجهزة إغلاق الجروح الحاجة إلى تقنيات فعالة من حيث القيود المفروضة عليها من التكلفة والكفاءة في التئام الجروح. ولقد كان هناك تطور وتوسع على نطاق كبير في أبحاث المواد والأعمال المتعلقة بالتطبيقات الطبية الحيوية.

حتى الآن، كانت الغرز والدبابيس هي الأدوات الرئيسية المستخدمة في صناعة الطب الحيوي. وتعتبر الغرز القابلة للامتصاص والتي تعتمد على البوليمر الحيوي مفضلة كثيراً على الخيوط غير القابلة للامتصاص. حيث تتحلل الغرز القابلة للامتصاص داخل الجسم عادة بمساعدة الإنزيمات المحللة للبروتين أو عن طريق التحلل المائي. وعلى الرغم من أنها متوافقة حيوياً، فهناك قيود في عدم وجود ما يكفي من مضادات الميكروبات، وتوصيل الأدوية المطلوبة للتطبيقات الطبية الحيوية.

وتهدف هذه الدراسة إلى تعديل كحول البولي فينيل القابل للامتصاص والشيتوزان وهو بوليمر حيوي ممزوج بـ GO كمادة نانوية والكيراتين كمصدر للبروتين (PVA /Cs/GO/ Ke) لدعم التأثير المضاد للميكروبات المحمي من الكائنات الحية الدقيقة المقاومة للأدوية المتعددة. وبالإضافة إلى ذلك، فهو يتمتع بمادة أكسيد الجرافين الموعودة لتعزيز الغرز قد يكون هذا بسبب توفير GO لمواقع كيميائية نشطة. ولقد تم استخدام GO أيضاً لتغليف الغرز الجراحية لتحسين وظائفها ومنع التصاق البكتيريا بها. لذلك، فهو نشاط جيد مضاد للميكروبات بحد ذاته أو في توصيل الدواء. الشيتوزان هو عامل مضاد للميكروبات مؤهل يمكن أن يعزى إلى طبيعته الكاتيونية ويقلل من محتوى الماء. وقد تم تحويل الخليط إلى ألياف نانوية بواسطة عملية الغزل الكهربائي. ثم تحويل الألياف إلى خيوط متعددة عن طريق آلة اللف.

وكشفت النتائج عن المزايا الرئيسية التي تتمثل فيما يلي أولاً في أن خيوط الألياف النانوية الممتصة ذات مضادات للميكروبات القوية ضد السلالات المسببة للأمراض، ثانياً، التحكم في وقت الامتصاص، ثالثاً، رخص المادة الحيوية وفي النهاية، فأنها ليست صديقة للبيئة فحسب بل و تعتمد على الشعر البشري.

التوافق الحيوي العالي للبوليمرات الحيوية،

التكلفة الاقتصادية للجراح،

التعقيم الذاتي دون المساس بسلامة المواد،

قابل للامتصاص بالمواقع النشطة بيولوجياً،

AS يتمتع بمقاومة البكتيريا.