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Review article

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Recent Trends in Catgut Therapy Using Textile Materials

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Abstract

The article comprehensively reviews recent developments in textile materials used for catgut embedding therapy applications. Polyglycolic acid (PGA) monofilament has been regarded as an excellent acupoint catgut embedding therapy (ACET) material because it offers numerous advantages, including easy accessibility and good forming and degradable properties. However, the poor hydrophilicity and cytocompatibility are the main disadvantages preventing it from having wider applications. Chitosan coating on acupoint catgut embedding therapy (ACET) is a critical issue in improving the comprehensive performances of embedding materials. However, the existing coating technologies have struggled to keep pace with both the academic study and industrial production. This work proposed a novel chitosan coating system consisted of pretreatment, spray-coating and rolling and drying parts. To evaluate the feasibility of this system, four types of monofilaments, namely polypropylene (PP), polylactic acid (PLA), polydioxanone (PDO) and polyglycolide acid (PGA), were adopted and their properties, such as swelling, compression and hydrophilicity, were also measured. Surgical site infections are liable to cause significant postoperative morbidity and increase health costs. However, polylactic acid and poly (D, L-lactide-co-glycolide) acupoint catgut-embedding therapy materials used to stimulate certain points for curing diseases are typically devoid of antibacterial activity. Novel polylactic acid and poly (D, L-lactide-coglycolide) embedding materials have been developed with effective antibacterial properties via chitosan-coating treatment that retained their inherent excellent characteristics. Acupoint catgut-embedding therapy (ACET) is considered one of the most promising technologies to replace traditional acupuncture therapy in clinical settings. Polyglycolic acid (PGA) monofilament shows great potential for ACET applications due to its advantages of good formability and biodegradability. The ideal embedding materials are required to have excellent swelling, mechanical, and antibacterial properties. For this paper we prepared two types of PGA monofilaments by first using the melt-spinning method and then applying chitosan onto the PGA monofilaments at three different concentrations.

Key Words: Catgut Embedding Therapy, Antibacterial Properties, Polyglycolic Acid, Swelling Behaviour, Chitosan, Polylactic Acid, Melt Spinning.

Introduction

Acupoint catgut-embedding therapy (ACET) research is a promising acupuncture treatment, characterized by the insertion of biodegradable materials into some parts of human body (1, 2). Once inserted, embedded materials in the acupoints will produce a series of stimulation effects to cure the disease concerned (3). Acupoint catgut-embedding therapy (ACET) is a medical procedure that implants biodegradable materials to stimulate some parts or acupoints of the human body, which has seen significant improvements since its introduction (4-6). It has the strong advantages reduced treatment times and a continuous stimulation effect compared with traditional acupuncture in the treatment of diseases such as juvenile myopia, obesity, and neuropathic (7-9). In the past decades, the prevalence of juvenile pseudomyopia has increased dramatically in the developed country or regions of East and Southeast Asia (10). Based on the World Health Organization, the rate of myopia remains high and growing, especially for young adults in these countries, they have gone from something like 20% myopia in the population to well over 80%, even spreading through the population when they get adult (11, 12). More than that, incident rate of this disease reaches up to 50% in China, and has become the most noteworthy threat for the young generation (13, 14).

Biodegradable Materials Coated and with Modified Surface:
In this field, ACET has attracted much attention because of its inherent advantages, such as its simple, effective, and lasting effects (15-17). However, there are still many potential problems for existing ACET materials. For example, surgical site infections (SSIs) are challenging complications after surgical procedures, which not only lengthen the time of hospitalization, but also increase the cost for patients, whereas embedding materials commonly used in clinics are typically short of antibacterial activity (18-20). Moreover, defects of poor mechanical properties,10 allergic reactions,11 and lack of functional characteristics have been cited as the major reasons for the decline in the production of ACET materials (21-24). Based on this, it is essential for researchers to study embedding materials for ACET applications. Biodegradable materials can be divided into natural and synthetic types according to their origin (25, 26). Catgut, a typical representative of natural embedding materials, has the advantages of good biodegradability, low cost, and easily accessibility (27). However, it is high risk to apply catgut material in the clinical treatment of allergy as it has variable quality. Synthetic embedding materials such as polylactic acid (PLA) and poly (D, L-lactide-co-glycolide) (PLGA), have multiple strengths such as excellent biodegradability and biocompatibility etc. (28-34). They have also been authorized by the US Food and Drug Administration to be used in clinical settings (35). Unfortunately, as reported by recent studies, degradation products of synthetic polymers such as PLA and PLGA generate acid products, which often cause inflammation when implanted (36). Hence, modifications are required to combine the advantages of natural and synthetic biodegradable materials, while avoiding their disadvantages. Chitosan is a linear, high molecular weight heteropolysaccharide, consisting of N-acetylglucosamine and N-glucosamine units (37, 38). With its abundant reserve, chitosan is the second most important natural polymer globally (the first is cellulose) and has been widely extracted from marine arthropods (prawns, crabs, shellfish, etc.) (39). As shown in our previous reports, chitosan-coating modification can be used to improve the surface roughness, mechanical properties, and biocompatibility of PLA- and PGA-embedding monofilaments (40, 41). Moreover, there are many applications of chitosan in the surface modification of textile materials for different purposes, including salt-free neutral dyeing of cotton, antibacterial cotton and wool, dyeing of wool, and antibacterial wound dressings (42-48). In recent years, applying chitosan coating on the surface of biodegradable material has become of interest. Zeng et al. studied the potential application of chitosan coating on three-dimensional porous PLA scaffold, proving this method to be useful in enhancing biocompatibility and cell adhesion properties (49). Han et al. investigated the effects of chitosan-coating technology on PLA films using a phase separation method (50). The results showed the hydrophilicity of composites was improved whereas the thermal stability was slightly reduced. The degradation rates could also be tuned in terms of structure through changing the pore sizes of PLA-based films. Moreover, the film showed strong antibacterial activity against Escherichia coli. Wang et al. fabricated biodegradable PLGA-chitosan core-shell nanocomposites with a narrow size distribution, and achieved good results in terms of drug-carrying capacity and sustained release performance (51). Feng et al. illustrated a new method for manufacturing PLA-aligned scaffolds modified by chitosan, and the viability, adhesion, length, and migration behaviors of osteoblasts in vitro was greatly improved (52). However, current studies are mainly concerned with coating chitosan on products such as scaffold, film, and nano composites, etc. and only examined the short-term modification effects. Few papers studied the effects of chitosan coating on linear materials including PLA monofilament and PLGA braiding threads. Moreover, there was lack of detailed research on in vitro comprehensive behaviors observed in the long term. Motivated by the surface modification of biodegradable materials using the chitosan-coating method to develop novel antibacterial embedding materials and retain their advantages, this paper first fabricated and compared four sets of PLA monofilaments and PLGA braiding threads. It then evaluated their comprehensive properties, including fundamental properties (surface morphology, weight and diameter changings, FT-IR analysis: For exploring the emergence of new functional groups of modified materials), mechanical properties (tensile, flexibility), and biocompatibility (cytotoxicity, antibacterial efficacy, and degradation). Finally, the feasibility of the chitosan-coating method on the PLA monofilament and PLGA braiding threads was evaluated based on these experimental results.

Novel PLA and PLGA embedding materials have been prepared using chitosan-coating treatment. To gain better comprehensive performance, their preparation processes were studied and a new one-dip one- rolling coating system was established. Characterizations such as structure, mechanical, and biocompatibility properties were measured to verify the feasibility of surface modification technology for developing novel antibacterial embedding materials. The following results could be concluded from this work (53).

1. By measuring structure characterizations, the surfaces of both the PLA and PLGA groups were shown to cover chitosan layers and some fragments; the CSPLGA group achieved more chitosan coating than the CS-PLA group due to its rougher surface structure. The FT-IR analysis results verify the existence of chitosan molecules and indicated that there were few new chemical bonds after surface modification.
2. Via the analysis of mechanical properties, the tensile properties and bending stiffness of PLA and PLGA groups were greatly improved after chitosan coating, and the CS-PLGA group presented the better mechanical performance than the CS-PLA group, caused by the larger amount of chitosan coating to which PLGA braiding threads adhered.
3. The biocompatibility evaluations demonstrated that all the samples were non-toxic and the modified PLA and PLGA groups demonstrated effective antibacterial efficacy against Staphylococcus aureus and Escherichia coli. Their weight-loss behaviour was delayed after coating treatment, which was beneficial for the lasting effect of embedding materials in the treatment period. Overall, the modified groups have alternative potential for product application in novel antibacterial ACET materials.
4. These findings suggested that chitosan coating was an easily operated and efficient method for PLA and PLGA groups to achieve requirements of antibacterial properties, while retaining the other advantages. More than that, the CS-PLGA group

with a larger amount of more chitosan coating seemed more suitable for ACET applications.

In summary, this study may inspire advancements in the design and fabrication of PLA and PLGA embedding materials with antibacterial activity to satisfy clinical applications.

Design of Innovative Polyglycolic Acid Monofilaments

Unfortunately, there is no agreement on the best material for use in ACET. The ideal embedding materials should satisfy several requirements, including excellent swelling, mechanical, and antibacterial properties. For swelling behaviour, the material could cause less surgical trauma due to its small diameter before implantation, and produce a larger acu point stimulation effect after expansion during treatment. It should have good mechanical properties for being embedded in the human body, and be able to bear the compression of muscle tissues (54). Antibacterial properties are essential in the prevention of post-surgical infections (55-57). Therefore, compromises must be made in selecting the embedding materials, and it is a difficult task for clinicians to weigh the advantages and disadvantages of the available embedding materials. Currently, the existing embedding materials can be broadly divided into natural and synthetic types according to their origins. It is challenging to use natural materials due to their unfavourable factors, including allergic reactions and variable quality (58, 59). Synthetic embedding materials have gradually become the dominant products in the ACET market. Among them, polyglycolic acid (PGA) offers many advantages – it is nontoxic, has good biodegradability, and good fiber-forming ability, for example – and has become a focal point for its utility in many biomedical materials (60, 61). However, using a large-diameter PGA embedding material not only brings a greater degree of acupoint stimulation and long-lasting effect in treatment, but also more thread-embedding needle and surgery trauma. Accordingly, patients choose to avoid ACET treatment due to the physical pain and psychological fear. Further, PGA material is difficult to use in ACET application due to its low stiffness (62-64). Hence, work is needed to improve the properties of PGA if it is to be the most promising embedding material. Chitosan is mostly obtained by deacetylation of chitin. It is a natural polymer rich in natural resources and has unique chemical and biological properties, such as excellent biocompatibility, biodegradability, antibacterial properties, and swelling behaviour (65, 66). Up to now, the chitosan coating method has attracted most attention in the tissue-engineering field. Niekraszewicz and colleagues20 used chitosan to modify polypropylene (PP) mesh and achieved good results in terms of mechanical and chemical properties; the biological purity was also improved (67). Vandevord and colleagues studied the potential application of chitosan coated on biomedical materials, proving this method to be useful in enhancing the biocompatibility of scaffolds. Dotto and colleagues investigated the effects of the chitosan coating method on biological films using diffusional models (68). The result showed its adsorption rate, antibacterial properties, and swelling behavior were greatly improved. Umair and colleagues reported that the PGA suture using N-halamine-based chitosan agents, its antibacterial efficacy was enhanced and could kill both E. coli and S. aureus bacteria within 15 min of contact time (69). However, most reports only used chitosan treatment on mesh, scaffold, film, etc. and regarded biocompatibility as the most important performance requirement. Few papers have focused on effects of monofilaments, and the application of the chitosan coating treatment is therefore worth further investigation.

In this study we aim to develop a novel antibacterial PGA monofilament for ACET applications using the chitosan dip-coating method. First, PGA polymer beads were adopted to spin into two types of monofilaments. Then, a chitosan coating agent at three different concentrations was coated on the monofilaments. The effects of chitosan coating on the properties, such as surface morphology, swelling, mechanical, FT-IR, antibacterial, and degradability properties were studied. The feasibility of the chitosan coating method was also evaluated according to the experimental results.

Two types of PGA monofilaments were spun from PGA polymer beads, and then treated with chitosan of different coating concentrations by the dip-coating method. The properties, including surface morphology, swelling, mechanical, FT-IR, antibacterial efficacy, cyto compatibility, and degradability properties, were tested to verify the feasibility of the chitosan coating technology for using in embedding materials (70).

The following can be concluded from the experimental results that.

1. Based on the analysis of SEM images, chitosan layers and fragments covered the surface of the PGA monofilaments, and the homogeneity of samples increased with chitosan coating concentration. Furthermore, 2-PGA monofilament showed a smoother surface than that of the 1-PGA monofilament at the same coating concentration.
2. By the measurement of swelling behavior, expansion rates of PGA monofilaments were greatly improved after chitosan dip-coating treatment, and PGA monofilaments treated with chitosan coating concentration of 3% (expansion rate: 1-PGA 3%¼98.32_4.24%, 2-PGA 3%¼123.05_2.87%) exhibited the most promising potential in the ACET applications.
3. The mechanical measurement revealed that the chitosan coating enhanced the tensile properties and stiffness of PGA monofilaments, and 2-PGA monofilaments with the larger diameters could adhere to more chitosan molecules and showed better mechanical properties than did the 1-PGA monofilaments. There were few interactions between the PGA monofilaments and chitosan molecules according to the FT-IR analysis. Antibacterial efficacy and cytocompatibility of the PGA monofilaments were also improved after the chitosan coating treatment. The degradation cycles of samples were delayed to different degrees for the protection of chitosan layers.
4. These findings suggest that the novel antibacterial PGA materials for ACET applications were successful developed by the chitosan dip-coating method, and the chitosan coating concentration of 3% should be selected as the optimum parameter for achieving the original design of this work.

Moreover, 2-PGA monofilaments with the larger diameters seem more suitable for ACET fields compared with 1-PGA monofilaments. This study may inspire advancements in the design and manufacture of PGA embedding materials with excellent comprehensive properties to satisfy clinical applications in further research.

Design of Polylactic Acid and Polyglycolic Acid Monofilaments

In the clinical areas, surgical is the most directly way for juvenile pseudomyopia, but the treatment is less effective, and it usually accompanies high risks (71). The drug therapy also cannot be the preferred method due to its shortcomings of toxic side effects, drug residual and resistance (72). Neither of these previous methods is ideal in the modern medicine field. Fortunately, Acupoint catgut embedding therapy (ACET), inserting biodegradable materials into some points or parts of human body, which has been regarded as the most promising technology to replace the traditional acupuncture therapy due to its advantages of convenient operation and lasting effect (73, 74). In the recent years, ACET field is moving towards more sophisticated applications with the addition of very specific high-end functionalities, such as antibacterial property, nanotechnology, drug release, etc. Reportedly, besides to the excellent biodegradability and biocompatibility, the ideal embedding materials should possess outstanding mechanical, biocompability and degradable properties (75). For example, embedding materials with a slow degradation behavior could produce long-term stimulation effect in the human body, and mechanical property is a physical characteristic that affects the smooth progress of surgical operation in the implanting process. However, the existing materials are still insufficient to meet the requirements of ACET application, and their preparation conditions need to be further studied. As such, some attempts should be carried out by the researchers to fabricate the embedding materials with excellent comprehensive properties (76, 77). Among biomedical polymers, polylactic acid (PLA) and polyglycolide acid (PGA) have drawn much attention because of the favorable formability and easy availability, and they are successful applied in clinical and certificated by the U.S. Food and Drug Administration (78, 80). In details, PLA is a kind of condensation polymer (aliphatic polyester), and it could be derived from completely renewable resources such as sugar cane and beet. Meanwhile, PLA monofilament possesses the high strength modulus and excellent biocompatibility, but its tenacity and impact resistance were unsatisfactory in the clinical practice, which indicates that PLA is strong and not flexibility enough (81, 82). In comparison of PLA, PGA has excellent physicochemical and mechanical properties due to its high regular crystal structure. Nevertheless, the poor machinability and swelling behaviour of PGA were undesirable as embedding materials (83, 84). Several reports of developing the ideal PLA and PGA embedding monofilaments by melt-spinning method have gradually attracted people's attention. The effects of melt spinning process on the structure of PGA monofilament were studied, and the results showed that PGA had a relatively small crystallinity without the drawing process, and the high speed spinning was not conducive to the degree of macromolecular orientation and the mechanical properties of PGA monofilaments (85). Moreover, PGA monofilament's mechanical and thermal properties were improved after the hot drawing and setting process. Furthermore, the spinning parameters were optimized according to the experimental results (86). The features of PLA and its polymers, quality control steps and the fabrication process of PLA were discussed, revealing that some factors such as impurities in the reagent lactic acid solutions and spinning conditions have directly influenced the properties of PLA, and different analyses for PLA should be highlighted (87, 88). Even though several studies have been conducted on the PLA and PGA monofilaments, but the effects of spinning parameters on the mechanical, in vitro properties of PLA and PGA embedding monofilaments have not been discussed and analyzed in details, and these performances were most noteworthy for the ideal ACET materials. Hence, it is worthy for a further study of their preparation process, which will also be conducive for our next surface modification research.

Fabrication has been done on different types of PLA and PGA monofilaments at different spinning parameters. Meanwhile, some theoretical calculations were applied to design the novel melt-spinning system and optimize the preparation process. Afterwards, the properties such as surface morphology, mechanical performances (tensile property and flexibility) and in vitro experiment (cell cytocompatibility and degradable properties) were tested and analyzed, and the feasibility of melt-spinning technology to prepare embedding monofilament was evaluated according to these experimental results (89).

PLA and PGA embedding monofilaments have been prepared by using different spinning conditions with a novel melt-spinning system. Both the PLA and PGA groups showed relatively smooth surfaces, and their diameters slightly decreased with the spinning parameters. The small spinning speed and the large drawing ratio were conducive to the tensile strength and bending stiffness of monofilaments, and PLA group exhibited better mechanical properties than that of PGA group. All the samples were nontoxicity and had the potential application on ACET fields. Through degradation property analysis, PLA group showed much slower degradation time than that of PGA group. PLA-2 and PGA-2 have the most suitable mechanical and degradation properties and could be prioritized as embedding materials. In sum, our data suggest that this melt-spinning system was feasible, and it was an easy-operated and efficient method for both the PLA and PGA groups to achieve ACET materials requirements of good mechanical and degradation properties.

Conclusion

To achieve more comprehensive properties, polylactic acid polymer chips and poly (D, Llactide- co-glycolide) multifilaments were adopted to fabricate embedding materials, and their preparation processes were studied. Meanwhile, a new one-dip-one-rolling coating system was designed and established in this work. Afterwards, characterizations such as fundamental properties, mechanical properties and biocompatibility were explored. The results showed that both the polylactic acid and poly (D, L-lactide-co-glycolide) groups with covered chitosan layers showed a different increase in weight and diameter. FT-IR analysis indicated that there were interactions between the pure embedding materials and chitosan molecules, the present of hydroxyl group (-OH) and some polar bonds such as (N-H) amide II and (C=O) amide I would be benefit for the hydrophilicity of modified materials. Chitosan-coated polylactic acid monofilaments in group 2 (breaking strength 31.26 cN/dtex, breaking elongation 34.82%) and chitosan-coated poly (D, Llactide- co-glycolide) braiding threads in group 2 (breaking strength 92.58 cN/dtex, breaking elongation 73.39%) showed better mechanical properties. Antibacterial efficacy and cell cytocompatibility of polylactic acid and poly (D, L-lactide-co-glycolide) groups were greatly enhanced after coating. The degradation behaviors were slowed down, providing a longer-lasting effect during the treatment process. In conclusion, the modified polylactic acid and poly (D, L-lactide-co-glycolide) embedding materials with good, comprehensive performance presented great potential in acupoint catgut-embedding therapy clinical applications.

Two types of PGA monofilaments have been prepared by first using the melt-spinning method and then applying chitosan onto the PGA monofilaments at three different concentrations. The effects of chitosan coating on the characteristics such as surface morphology, swelling, mechanical, FT-IR, antibacterial, and degradable properties of monofilaments were explored. The results show that chitosan layers and fragments were distributed uniformly on the surface of PGA monofilaments. The expansion rate of samples increased with the concentration of the chitosan coat, and samples 1-PGA 3% (98.32%) and 2-PGA 3% (123.05%) exhibited the best swelling behavior. Mechanical properties of PGA monofilaments were greatly improved after chitosan coating. There were few interactions between PGA monofilaments and chitosan molecules according to the FT-IR analysis. Antibacterial efficacy and cytocompatibility of PGA monofilaments were greatly enhanced by the chitosan coating treatment. The degradation time was delayed due to the protective function of the chitosan layers, producing a more lasting effect in the treatment process. In conclusion, the novel PGA embedding monofilaments were successfully developed and a chitosan coating concentration of 3% was chosen as the optimum parameter.

Novel melt-spinning system has been designed to produce different PLA and PGA monofilaments, and effects of spinning conditions on their comprehensive performances were characterized. The results showed that samples had the smooth surfaces and diameters decreased with spinning parameters. PLA-2 (Strength 78.63 cN; Elongation 57.28%) and PGA-2 (Strength 57.34 cN; Elongation 18.42%) perform the best mechanical properties. All the samples were non-toxicity, and PLA group degraded much slower than that of PGA group. In conclusion, this melt-spinning system showed potential in ACET application the optimum parameters were proved to have the higher prospect.

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