

Some Advances in Textile Materials Used in Acupoint Catgut Therapy Application

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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.

Keywords: Polyglycolic Acid; Acupoint Catgut Embedding Therapy; Cytocompatibility; Chitosan; Coating System; Swelling

Introduction

Recently, acupoint catgut embedding therapy (ACET) has gained wide recognition as an advanced acupuncture method for the treatment for various diseases, such as obesity, neuropathic pain, juvenile pseudo myopia, etc [1-3]. The mechanism of this therapy is to implant biodegradable materials into some parts of the human body, further providing a sustained stimulation for meridian acupoints [4,5]. As such, embedding materials have successfully replaced acupuncture needles in the treatment process, as they avoid many disadvantages of traditional approaches, including short-term efficacy, frequent treatment and immune reactions [6]. Unfortunately, the difficulty of design and fabrication of the ideal biodegradable embedding materials has limited the application of ACET in clinical applications.

Acupoint catgut embedding therapy (ACET) is widely used in the clinical field as advanced acupuncture for increasing unmet medical needs. The treatment process can be defined as inserting biodegradable materials into some points or parts of human body for curing diseases [7-9]. Generally, suture material is buried in a thread-embedding needle and directly acts as a substitute for embedding material in the clinical field, causing varied problems, such as contradiction between the lasting effect and large trauma, an incompatible degradation time with physical recovery, etc [10,11]. Thus, it is essential for the clinical researcher to weight the usage of embedding materials based on the health of different patients. In addition to the good biodegradability and biocompatibility, the ideal embedding materials are required to possess many other advantages [12,13].

Surface modification of polyglycolic acid mono filaments by combination of hydrogen peroxide and ultrasound surface modification

Polyglycolic acid (PGA) has held great promise for various biomedical applications in human tissue due to its advantages of being easily available, low cost and having excellent biodegradability and biocompatibility [14]. Moreover, unlike natural biodegradable materials that easily cause infections, PGA monofilaments prepared by melt-spinning technology for ACET materials have exhibited sufficient supply, good formation capacity and versatility in terms of surface functionalization [15,16]. Based on these attractive functional characteristics, the PGA monofilament is not only certified by the US Food and Drug Administration (FDA), but also considered to be a highly promising biodegradable material for ideal ACET material preparation [17,18]. However, the PGA monofilament belongs to the category of linear material, not having a tortuous porous structure and large surface area. These features are believed to be the main contributors to the unfavorable characteristics [19]. We thus expected that the PGA monofilament can be surface modified to enhance its surface roughness and complexity, consequently improving the hydrophilicity and cytocompatibility properties.

In the past decade, there has been an array of surface modification technologies, including surface coating, plasma modification, iCVD, ion beam injection, surface graft polymerization, etc., applied to PGA materials. For example, with the use of the coating method, collagen-coated PGA conduits were produced and applied in treating facial nerve paralysis in rats. The results showed that the modified PGA conduits presented better histology and physiology properties than that of the unmodified PGA conduits [20]. The surfaces of PGA nanofibrous scaffolds were chemically modified using oxygen plasma treatment and *in situ* grafting of hydrophilic acrylic acid (AA), and better cell attachment and proliferation *in vitro* for the modified PGA were proven [21]. By initiating the iCVD technology, a cellulose nanofibril (CNF) and PGA-containing multilayer film was developed, and it showed promising oxygen barrier improvements at different humidities [22]. Even though much research has been conducted on the surface modification of biomaterials, the improvement of hydrophilicity and cytocompatibility properties is still relatively limited, and it is also a challenge to retain the original excellent characteristics of biomaterials after modification. As a popular approach, the mechanism of ultrasound treatment was used due to the phenomenon of cavitation, which involves nucleation and the behaviour of bubbles in the liquid is suitable [23,24].

Meanwhile, high-energy phenomena, including elevated temperature and pressure and electrical discharges, will also occur during the treatment process [25,26]. Compared with conventional chemical modifications, ultrasound treatment has been regarded as a simple and efficient surface modification technology for various models of textile fibers. Wang, *et al.* applied ultrasound treatment on the surface modification of PGA and poly lactic-co-glycolic acid (PLGA) fibers [27]. The results showed that the hydrophilicity and cytocompatibility of modified fibers were greatly enhanced, while their mechanical properties changed little. For example, polyester fiber was treated with ultrasound modification, and the result showed that its wettability was enhanced via observing the decrease of the contact angle at each testing point [28]. In addition, combining ultrasound treatment with coating technology was also considered to be useful in surface modification applications. Savelyeva, *et al.* coated a calcium carbonate (CaCO_3) layer, which was composed of vaterite microparticles and their aggregates, on the surface of poly(*e*-caprolactone) (PCL) fibers under ultrasound treatment, and a porous structure, good biological activity, cell adhesion and spreading properties were observed [29]. More than that, numerous reports have focused on using ultrasound treatment for the surface modification of fibers such as wool, lignocellulosic, aramid, etc. [30-32]. Although ultrasound treatment has been considered as one of the most promising surface-modified technologies for textile materials, the effects of ultrasound frequency on biodegradable materials have been little examined.

The goal of this study was to fabricate novel PGA embedding materials with good comprehensive properties for ACET clinical applications. For evaluating the effects of diameters for PGA monofilaments on the surface modification, two types of PGA were firstly produced from their polymer chips using the melt-spinning method. Subsequently, hydrogen peroxide solution was adopted as a modified agent to improve the roughness of the prepared monofilaments at three different ultrasound frequencies (45, 60 and 75 KHz). Finally, the modified

PGA monofilaments were deeply characterized to evaluate the structure characterizations (surface morphology, weights and diameters, Fourier transform infrared spectroscopy (FT-IR) analysis and hydrophilicity), mechanical properties (tensile property, swelling behavior and flexibility) and *in vitro* properties (cytotoxicity, cell attachment and cell morphology) [33]. The ultrasound/H₂O₂ surface-modified PGA monofilaments may have great potential for practical applications. They may provide a good alternative for use as ACET material. This paper firstly fabricated PGA monofilaments with different diameters using melt-spinning technology, then adopted ultrasound surface modification for these prepared monofilaments. Meanwhile, different ultrasound frequencies were applied and investigated in the modification process. The following conclusions were established through this work:

- Via the analysis of structure characterizations, all the modified samples presented rough surfaces, while the unmodified samples showed relatively smooth surfaces and small weights and diameters. The FT-IR analysis indicated that some formed polar groups, including hydrophilic hydroxyls and carboxyls on the terminal of molecules, which promoted the affinity with water molecules for modified PGA monofilaments;
- By the measurement of mechanical properties, the ultrasound treatment was proven to enhance the swelling behavior of the 1-PGA and 2-PGA groups, and slightly decrease the tensile strength and bending stiffness, but the difference was not obvious;
- The *in vitro* experiments showed that all the samples were non-toxic with the more than 75% cell viability, and the modified samples presented a much larger cell attachment ratio than that of the unmodified samples. In detail, the 2-PGA group showed a better cell attachment ability at the same ultrasound modified frequency due to its larger diameter. Hence, the 2-PGA group showed better potential for clinical applications;
- These results suggested that we have demonstrated a successful surface modification of PGA monofilaments for improving hydrophilicity and cytocompatibility. Such excellent comprehensive properties make the modified PGA embedding materials good candidates for ACET applications;
- In summary, this study may inspire advancements in the design and manufacturing of novel PGA monofilaments to satisfy clinical requirements.

Innovative design of chitosan coating system

They should provide enough mechanical properties to be implanted into the body and support peripheral nerve tissue [34]. Through good swelling behavior they are able to produce enough stimulus degree *in vivo*, yet retain a relatively small dimension *in vitro* and avoid the significant trauma [35]. Favorable hydrophilicity is also important for embedding material to adhere cells and work well in the human body [36]. Hence, several attempts must be carried out to modify the properties of embedding materials to meet the ideal standard. Chitosan is widely used in numerous fields as a natural macromolecular material with excellent properties, such as biocompatibility, biodegradability, nontoxicity, antimicrobial activity, swelling behavior, etc [37,38]. In recent years, it has provided a new insight into the ACET field in clinical applications to applying chitosan for the improvement of comprehensive performances of biodegradable materials [39,40]. For example, the chitosan-coated scaffold presented a superior biocompatibility, biodegradability and a slow biodegradation, which retarded bone regeneration [41]. The chitosan-containing liposome could be used as antibacterial drug delivery for the treatment of vaginal infections and achieve good effect [41]. In addition, the degradation time of chitosan/sodium alginate hydrogels could be delayed with the increase of chitosan coating weight [42]. Actually, the treatment effect of embedding material was better with the increase of chitosan molecules and acting time in the human body. Accordingly, a series of coating techniques was developed for the deposition of chitosan layers on substrates by the following methods: wet chemical processes, spin coating, spray coating, roll-to-roll coating and dip-coating [43-46]. Up to now, solution coating technology is reported to deposit aqueous-based liquid coating agent onto any substrate, including metallic, ceramic, polymer film, fibrous material, etc [47-50]. Hence, this method has been widely used in numerous industrial

fields due to its advantages of easy operation, convenient for use and comprehensive functions [51,52]. Ceratti, *et al.* developed a new, simple bi-phasic coating method to improve the utilization ratio of coating solutions and could deposit small amounts of solution onto large surfaces [53]. Lii, *et al.* indicated that turbostratic boron nitride films could be deposited onto carbon fibers and graphite substrates by coating in methanolic boric acid and urea solutions [54]. Faustini, *et al.* fabricated a unique functional device that not only controlled the gradient of properties by the coating process in acceleration mode, but also prepared thin films with “on-demand” thickness graded profiles in an easy and versatile way [55,56]. Based on this, embedding material is proposed to be processed by the chitosan coating method, combining advantages of chitosan swelling behavior and coating technology’s facile economical properties, while attempting to make up the defects of the existing embedding materials.

However, these researches offered more focus on products such as surgical sutures, films, pelvic floor patches, nerve scaffolds, etc. Few papers have studied and analyzed the application of chitosan coating technology on monofilaments. Also, a single immersion method was insufficient to achieve the coating amount required in experiments. This paper designed a novel chitosan coating system for biomedical materials and optimized it by the theoretical analysis of the coating device [57]. To verify the feasibility of this coating system, four types of embedding monofilaments, namely polypropylene (PP), polylactic acid (PLA), polydioxanone (PDO) and polyglycolide acid (PGA), were adopted in this work, and then their properties, such as swelling, compression and hydrophilicity, were compared and discussed to achieve the optimum parameters. In the present work, the chitosan coating system for biomedical materials designed and optimized by the analysis of theoretical calculation was successfully developed, which offered a potential method to fabricate the ideal embedding monofilaments. The four types of monofilaments, namely PP, PLA, PDO and PGA, were compared with each other for the different testing properties, and verify the feasibility of this coating system. Testing variables were also optimized by estimating the testing properties of samples. The following conclusions were established through this study:

- The relationship between the coating concentration and testing properties showed the following. The coating concentration of 3% (v/v) was preferential in the experiment, because the testing properties, including swelling behavior, compression and hydrophilicity properties, were improved with the increase of coating concentration (< 3%), but they changed little under the coating concentrations of 4% and 5%.
- The rotational speed of 2.12 m/min should be selected as the optimum parameter. Actually, a high rotational speed of monofilaments made it difficult for them to be coated with chitosan molecules, further causing a lower chitosan coating amount and the lower than expected testing properties of samples.
- With regard to the testing results of samples, contact force of 0.1 cN was the most suitable in the measurement.
- The large contact force not only reduced the number of chitosan molecules that covered the surface of monofilaments, but also may cause some damages to the performances of samples. In contrast, a small contact force contributed to obtaining better swelling, compression and hydrophilicity properties.
- The changing properties of different samples (PP, PLA, PDO and PGA) under the variables, such as coating concentration, rotational speed and contact force, proved that the theoretical analysis and calculations were accurate and workable. Furthermore, this spray-coating and rolling system had a potential application in fabricating the ideal embedding monofilament to satisfy clinical needs.

In summary, these results suggested that this spray coating and rolling system was an easily operated and efficient method for embedding materials to achieve comprehensive performance, so it could be applied for ACET applications in our next study.

Conclusion

In this communication, two types of PGA monofilaments were first fabricated from their polymer chips, followed by ultrasound/H₂O₂ combined surface modification at 1:1 (V/V), and different ultrasound frequencies (45, 60 and 75 KHz) were explored. The modified PGA monofilaments were fully characterized with respect to structure characterizations (surface morphology, weights and diameters, Fourier transform infrared spectroscopy (FT-IR) analysis and hydrophilicity), mechanical properties (tensile property, swelling behavior and flexibility) and *in vitro* properties (cytotoxicity, cell attachment and cell morphology). The results showed that the PGA monofilaments after modification would become coarser, with larger weights and diameters. Samples 1-PGA 75 and 2-PGA 75 exhibited the smallest contact angles at 70.51° and 62.84°, respectively. The FT-IR analysis results confirmed that some polar groups emerged, promoting the hydrophilicity of PGA monofilaments. The swelling behavior of monofilaments was enhanced, while tensile and bending stiffness values slightly decreased. All the prepared samples presented no toxicity, and the cell attachment ratio (cultured for 48h) of PGA monofilaments had been greatly improved after modification. These findings present important clinical implications in the ACET materials manufacturing process and warrant further study to develop new PGA embedding materials with outstanding clinical efficacy. Meanwhile, several numerical calculations and analyses were conducted to select and optimize the testing variables. The results showed that the coating concentration of 3% (v/v) was selected as the most suitable parameter; samples with the rotational speed of 2.12 m/min were demonstrated to have regular swelling behavior: PP < PLA < PDO < PGA; testing properties of samples were closer to the expected results regarding the decrease of contact force, so 0.1 cN was selected as the optimum parameter. In summary, the coating system was successfully developed and has a potential application for fabricating ideal embedding materials to satisfy clinical needs.

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