Stretchable Conductive Fabric for Electrotherapy

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ABSTRACT: Changes in conductivity with repeated fabric extension were investigated to improve the properties of conductive electrode pad material used for electrotherapy when it is subjected to various movement of human body. Highly stretchable and conductive fabrics were prepared by *in situ* chemical polymerization of polypyrrole on nylon–spandex stretch fabric in aqueous solutions with 0.5 M pyrrole, 1.165 M $FeCl_3$, and 0.165 M benzenesulfonic acid at 5°C for 1 h. Performance of prepared stretchable conductive fabric was evaluated in terms of conductivity changes as a function of tensile strain, repeated extension, and current application time. As the degree of extension increased, the conductivity increased and leveled off when the fabric was subjected to 60% extension. The number of fiber contacts in nylon–spandex fabric with electrode increased as the applied extension increased. However, the conductivity of the composite decreased under excessive extension over 60% since the intrinsic elasticity of fabric became gradually reduced. Generally, the fabric conductivity decreased as the number of extension cycles increased. However, the fabric conductivity was well maintained after repeated extension over 30 cycles at 40% extension. In addition, it was found that the effect of charging during the electrotherapy treatment on a current flow through prepared electrode pad was negligible. © 2003 Wiley Periodicals, Inc. J Appl Polym Sci 88: 1225–1229, 2003

Key words: stretchable conductive fabric; polypyrrole; extension; electrotherapy

INTRODUCTION

Recently, electrotherapy using electric pulses is increasingly being used in physiotherapy and rehabilitation to reduce pain, enhance healing, and improve patient's mobility.[1–3] However, in the case of general electrotherapy, there are problems with the surface electrode placed on the skin or needle electrode inserted into the living body, such as its conductivity and the method of application. Therefore, the interest in manufacturing pad electrode materials with improved conductive and physical properties has grown.

Conductive materials used for electric stimulation electrodes are usually manufactured by the posttreatment of fabric or addition of conductive fillers to the fiber on the spinning step. In particular, posttreatment has been used mainly because it is easy to apply to the fabric finishing process. Posttreatment includes many techniques, such as metal coating, resin processing with conductive materials, metal plating, and coating with polymer containing a conjugated π-electron backbone. Among the techniques, conductive polymer coating imparts flexibility and durability to a fabric. Therefore, the pad electrode materials can be prepared by using the π-conjugated polymer such as polypyrrole, polyaniline, and polythiophene on fabrics.[4] Among these, polypyrrole has excellent environmental stability, facile synthesis, and higher conductivity than many other conductive polymers.[5–7] Pyrrole was first chemically polymerized in 1916 by the oxidation of pyrrole with water to give a powder state known as pyrrole black. However, the application of pyrrole black is limited because it is insoluble and infusible. Therefore, various methods for the application of conducting polymer were developed in the form of conductive fabric and blend-spun fiber.[8]

In this research, polypyrrole was *in situ* polymerized chemically on nylon–spandex stretch fabric to complement the difficulties in processing and to provide flexibility, elasticity, and ideal conformability to pad materials. Changes in conductivity with repeated extension were investigated to improve the properties of conductive electrode pad materials to be adaptable for human body movement during electrotherapy treatment. In addition, changes in conductivity with the volume of the direct current were observed to ascertain the effect of generated heat from a long period of treatment current for electrotherapy.[9–11]

EXPERIMENTAL

Materials

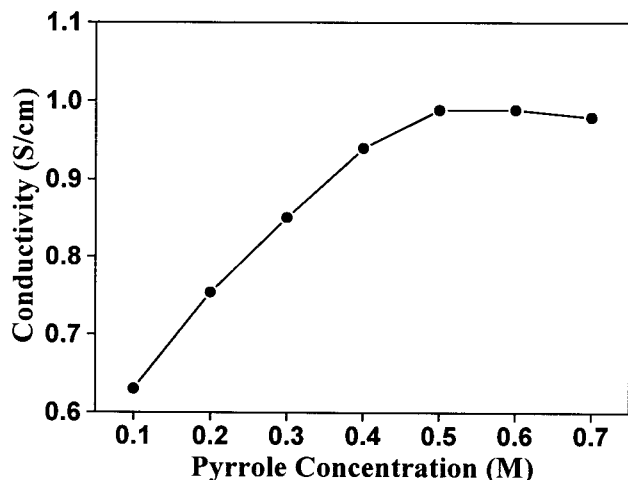Nylon–spandex (95:5) stretch fabrics (140 denier, warp 184, weft 256 in 25 cm^2; twill weave, Taewoo Fabrics)

Figure 1 Change of conductivity with pyrrole concentration.

Figure 2 Stress–strain curve of nylon–spandex fabric.

were washed with aqueous solution of scouring agent and sodium hydroxide for 1 h to remove spinning oil used in weaving process. Then the fabrics were completely rinsed and dried before use. Pyrrole (Aldrich) was used after vacuum distillation. Ferric chloride anhydrous (FeCl$_3$; Duksan Pure Chemical) and benzenesulfonic acid (BSA; Lancaster Synthesis) were used without further purification.

Preparation

Conductive fabrics were prepared by *in situ* chemical polymerization of pyrrole to retain elasticity of stretch fabrics in the presence of textile substrates. Pyrrole was introduced into an aqueous solution of BSA. After monomer was completely stirred up, the fabric was immersed in the solution at room temperature for 1 h.[12,13] During diffusion step, ultrasonic treatment (Bransonic 3510R-DTH, 42 kHz) was employed for 20 min to improve the degree of monomer diffusion into the dense fabric. Ferric chloride solution containing BSA was added to diffusion bath to initiate polymerization.[14] Polymerization was carried out at 5°C for 1 h. The optimum molar ratio of monomer, oxidant, and dopant was determined to be 1:2.33:0.33.[15,16] The conductive fabrics were well washed with flowing

water and dried in vacuo at 40°C for 24 h. Polymerization occurred on the surface of the substrate, which each fiber was individually coated with polypyrrole.

Characterization

Conductivity was measured with resistance tester (Sanwa) and digital multimeter (Keithley 238, Keithley Instruments). Fabrics were dried sufficiently before measuring conductivity. The change of the conductivity with extension was measured as a function of strain at substrate. The changes of mechanical properties of the conductive fabrics were investigated with tensile testing machine (UTM Instron 4465, Instron) by standard test methods for coated fabric (ASTM D 751-95).[17] The standard condition for tensile testing of coated fabrics was RH 65% and 25°C. The morphologies of each fiber with various extensions were observed by FE-SEM (field emission scanning electron microscopy; JEOL JSM-6330F, Japan).

TABLE I
Optimum Polymerization condition

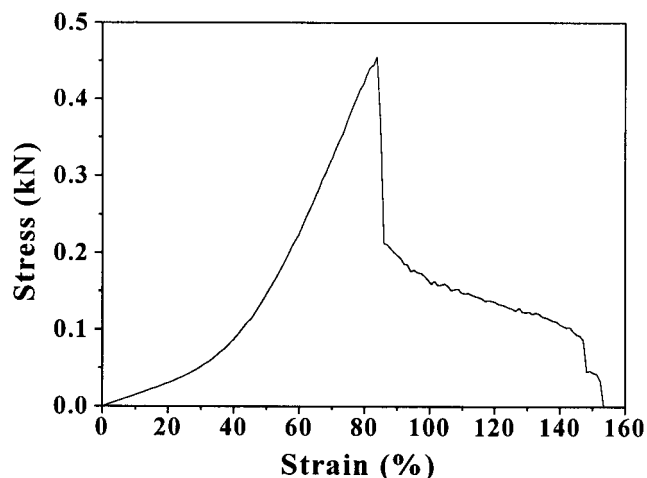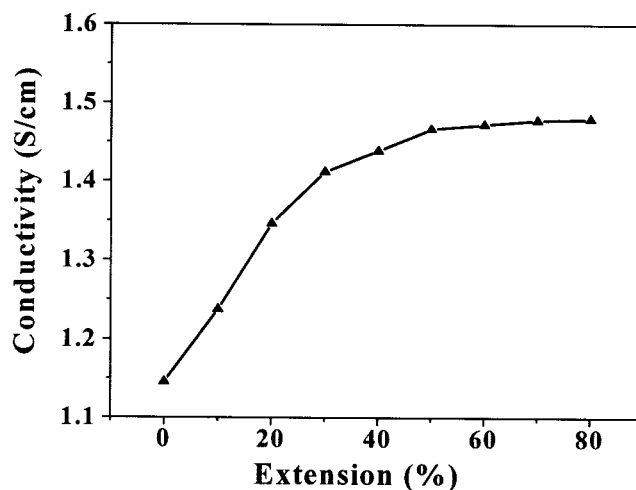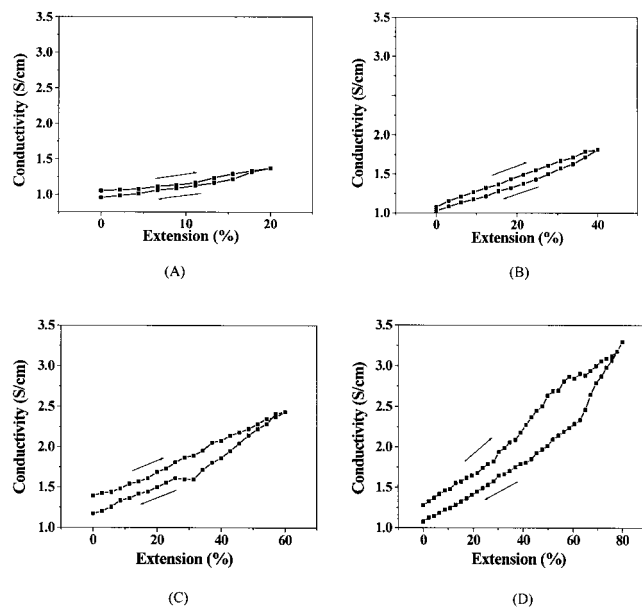| Factor	Optimum condition
Pyrrole concentration	0.5 M
Diffusion of monomer	At room temperature for 1 h (20-min ultrasonic treatment)
FeCl$_3$/pyrrole (molar ratio)	2.33
BSA/pyrrole (molar ratio)	0.33
Polymerization temperature	5°C

Figure 3 Change of conductivity with extension.

Figure 4 Conductivity changes during stretching and relaxation test: (A) 20%, (B) 40%, (C) 60%, and (D) 80% extension.

RESULTS AND DISCUSSION

Optimum polymerization condition

Conductive fabrics were prepared by *in situ* chemical polymerization with pyrrole monomer, oxidant, and dopant in order to prevent reduction of the substrate's elasticity. From our previous work, it was found that immersing the fabric in an aqueous acid solution of pyrrole and initiating polymerization by mixing the prepared oxidant and dopant solution could easily produce highly conductive fabric. The optimum molar ratio of pyrrole:oxidant:dopant was determined to be 1:2.33:0.33.[15,16] Thus, by fixing the molar ratio of pyrrole:oxidant:dopant, the experiment was carried out with increasing pyrrole concentration to determine optimum polymerization condition. The change of conductivity of fabric with different pyrrole concentration is shown in Figure 1. As the pyrrole concentration was increased up to 0.5 M, the fabric conductivity was increased and then leveled off. Therefore, the concentration of pyrrole was fixed at 0.5 M through this study. With BSA dopant, fabric conductivity could reach to approximately 1 S cm^{-1}. The optimum polymerization condition is given in Table I.

Conductivity changes with extension

Mechanical property of the fabric was measured with Instron 4465 by the grab method. The change of tensile stress with strain is shown in Figure 2. From the tensile test data, it was found that nylon–spandex core-spun yarns were decrimped when the fabric was stretched up to 20% extension, and nylon covering

TABLE II
Recovery Properties of Electroconductive Fabrics

	Residual strain (%)		Fractional elastic recovery with extension	
	1 cycle	10 cycles	1 cycle	10 cycles
Hysteresis at 20% extension	1.7193	2.1431	0.9143	0.8932
Hysteresis at 40% extension	9.4235	11.5681	0.7696	0.7172
Hysteresis at 60% extension	14.1165	19.4281	0.7681	0.6809

fibers were detwisted and extended as the level of extension was further increased. Finally, they began to break at 60% extension. Spandex monofilament used for core yarn was elongated up to nearly 80% and was broken beyond this point. Accordingly, the substrate started to break with over 80% extension. Therefore, extension limit was determined to be 80%.

A resistance change caused by the applied deformation was measured using resistance tester. The conductivity of the fabric at different elongation level has been calculated. Generally, the measured fabric conductivity values increased as the fabric was stretched up to 50% extension and then leveled off as shown in Figure 3. Initial increase in conductivity is obtained by the fact that the surface interval between two electrode probes was reduced by decrimping and detwisting of covering yarns with fabric extension. Moreover, it is probably due to a progressive increase in the number of contacts and the area contacting the bundles of fibers since fibers are closely rearranged as the applied load increased.

Additional experiments were performed at 20%, 40%, 60%, and 80% extension levels to identify con-
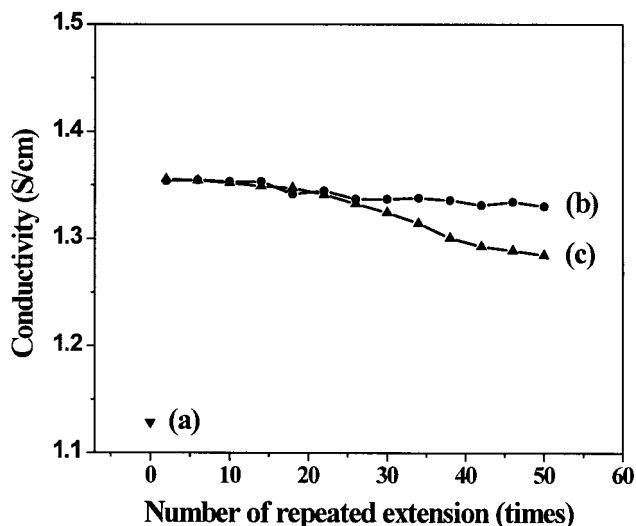

Figure 5 Change of conductivity with repeated extension at various levels: (a) initial conductivity, (b) 40%, and (c) 60%.
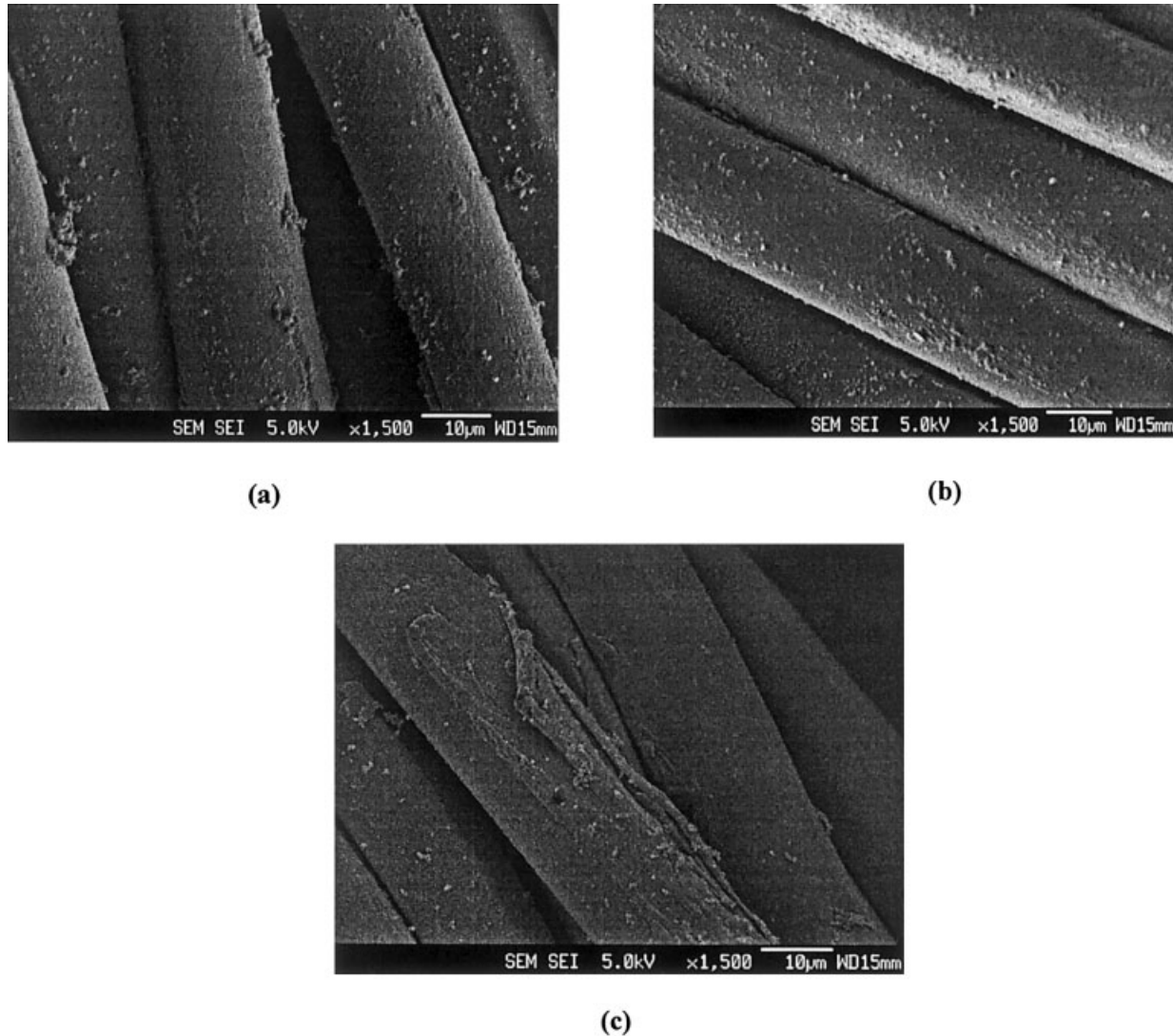
(a)

(b)

(c)

Figure 6 FE-SEM microphotographs of the nylon fibers used as covering fibers with magnification of 1,500: (a) before repeated extension, (b) after repeated extension over 30 cycles at 40% strain, and (c) after repeated extension over 30 cycles at 60% strain.

ductivity changes during stretching and relaxing of the fabric. In general, the fabric conductivity during relaxation test is lower than that during the stretching test, since fabrics are impaired during stretching. Therefore, conductivity hysteresis arises. Figure 4 shows the changes of conductivity with various levels of extension. Difference between the conductivity values on stretching and relaxing increased as the extension increased. As a result, it was found that the conductive fabric was mechanically stable up to approximately 40% extension, but hysteresis increased significantly over 60% extension where the covering fibers started to break.

Conductivity changes with repeated extension

From the tensile properties of conductive fabric as shown in Table II, it was expected that the fabric

elasticity was hardly influenced by 20% extension, but it would probably be lost at more than 40% strain level. Therefore, repeated stretching and relaxing tests were performed at 40% and 60% extension level because it was important to maintain the fabric conductivity after multiple usage of wearable electrode.

In general, conductivity of composites was not significantly decreased and well maintained when it was subjected to 40% extension as shown in Figure 5. However, nylon fibers used as covering yarns began to be extremely extended at 60% extension level, and damages on the surface interval started to appear. Therefore, the generated damages on the surface interval between two electrodes and separation of polypyrrole due to repeated excessive extension cause reduction of fabric conductivity and their durability. It was further confirmed by the elastic recovery data and FE-SEM microphotograph as shown in Figure 6. As
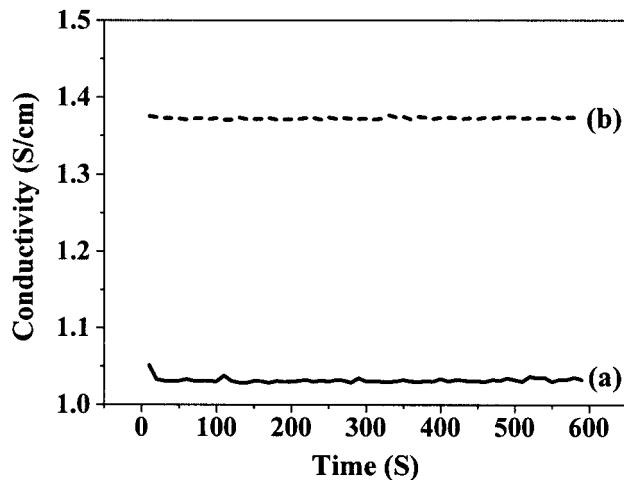
Figure 7 Change of conductivity with time on different conductivity measuring current: (a) 10 mA and (b) 20 mA.

the percentage of repeated extension increased up to 80%, the polypyrrole layer was separated from the fiber, reducing the fabric conductivity.

Conductivity changes with time

The conductivity variation in measuring current of conductive fabrics is shown in Figure 7. Currents for medical use are the direct current, the induction current, and the alternating current. A direct current is a current whose electricity possesses a fixed current flows in the constant direction using a dry cell, a storage battery, and so on. The induction current is a changeable current that was electrified with induction coil. The direct current of medical use is electrified to nerve or muscle with the range of voltage from 4.5 to 45 V and the current from 0.1 to 10 mA and the induction current for medical use has the current from 2 to 10 mA. Therefore, the experiment was performed at 10 and 20 mA to observe the variation of times of current volume since the ranges of the direct current used for medical application were generally from 0.1 to 10 mA. The conductivity did not change with time over 600 sec at the same current. During electrotherapy, therefore, the effect of prolonged current apply on a electrical resistance developed on the substrate can be negligible.

CONCLUSIONS

Changes in conductivity with extension and repeated extension were investigated to improve the properties of electrode pad material used for electrotherapy.

Polypyrrole was *in situ* polymerized chemically on nylon–spandex stretch fabric in order to provide flexibility and elasticity. Under the optimum reaction conditions, pyrrole of 0.5 M was used, since extensive monomer diffusion reduced physical properties of substrate without further increase in conductivity.

Generally, the conductivity of extended fabrics was continuously increased as the fabric was extended up to 60% strain while covering fibers were decrimped and detwisted. However, the fabric conductivity was decreased under excessive extension over 60% since the intrinsic elasticity of fabric became gradually reduced.

As the number of repeated extension cycles was increased, the fabric conductivity was hardly decreased and well maintained at 40% extension. On the other hand, the conductivity was decreased with repeated extension of more than 30 cycles at 60% extension rate due to generated defects on the surface interval between two electrodes. In addition, it was found that the effect of charging during the electrotherapy treatment on current flow through prepared electrode pad was negligible.

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