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Research Article

Application of PEDOT:PSS Conductive Polymer to Enhance the Conductivity of Natural Leather: Retanning Process

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Leather is a natural material, made of collagen fiber bundles used to produce diverse types of products such as gloves and shoes due to durability, viscoelasticity, and strength. However, there is a limitation to using it for smart products due to electrical insulator properties and it needs conductivity properties to use for smart products. This study showed the new method of increasing the conductivity of goat glove leather by using poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS), dimethyl sulfoxide (DMSO) as an enhancement reaction during the retanning process after leather acid dyeing. Analyze the properties of experimentally and conventionally treated leathers such as Fourier-transform infrared spectroscopy, tensile strength, color fastness, water absorption test, organoleptic, and electron microscopic analysis. The current study found that the treated leathers with 0.5 M PEDOT:PSS, 0.3 M DMSO, and acid dyestuff shows a maximum conductivity of 8.0 S/cm and can be used for making conductive gloves for operating touch-screen devices. Generally, it was found that acid-dyed leather samples treated by PEDOT:PSS with DMSO are more highly conductive than basic dyes and conductive polymers, which are applied on resining finishing. The study also found that experimented with conductive polymer leather processing has 38.4 kg/mm, 257.2% elongation, and 36.4 N tear strength, which are better than conventionally treated leather.

1. Introduction

Leather is a strong, flexible, and durable material obtained by the chemical treatment of animal skins and hides to prevent decay and retanning is one of the processing steps in leather manufacture to improve leather fullness and some physical properties [1, 2]. Nowadays, leather materials are dependent on revealing superfluous functionalities other than comfort and durability by modifying them using electrical and computer applications [3]. Leather is nonconductive material that makes it not to be used in smart product development [4]. The average surface electrical resistance, exhibited by human skin, is estimated to be around $10^5\ \Omega$, while leather surface resistance normally varies from about 10^9–$10^{10}\ \Omega$, depending on the kind of chemicals and technologies used in leather production [5, 6].

Furthermore, a disabled person who has lost a finger can use this conductive leather with a proper design to operate a smart touch screen device [7–9], for example, smartphones, tablets, laptops, and other mobile devices. In the harsh winter months, a glove made from highly conductive leather provides accurate control over touch-screen devices without any need to remove the gloves. To function touch-screen gadgets comfortably in the cold season, conductive leather is in high demand. Furthermore, a disabled person who has lost a finger in another circumstance can use this conductive leather with a proper design. Conductive leather can also be used in the creation of smart garments. Commercial techniques include applying conductive agents such as carbon black, carbon nanotubes, graphite, silver, copper, gold, and other metals to the surface of the leather during finishing, embroidering conductive thread on the fingers and thumbs, or knitting a conductive complex on the entire glove to pass electrical impulses to touch screens [10]. Previously one studied leather electrically conductive treatment by coating leather with the conductive polymer during leather finishing stages. However, the conductivity nature of leather was not stayed for a long duration due to noninteraction with fiber leather rather than a surface coating.

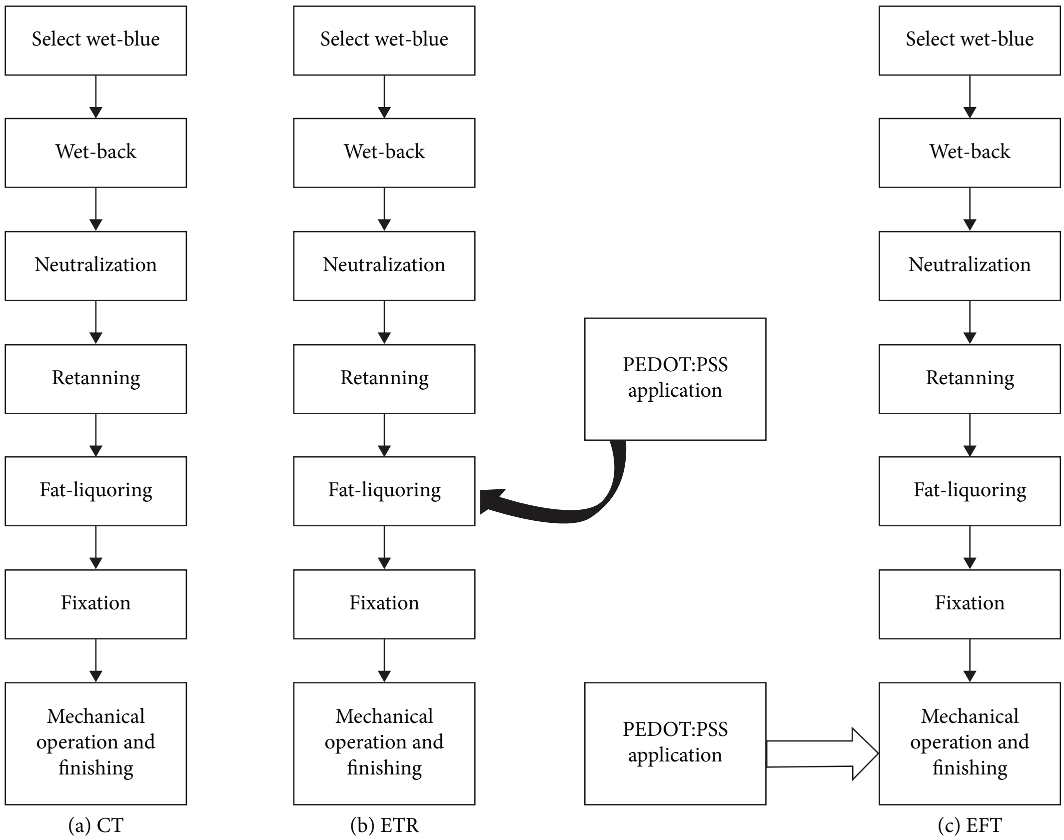


Figure 1: Study design.

Poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) is a known material to possess high conductivity [11], easy synthesis, good environmental stability, and less toxicity [12, 13]. It does not affect comfort too much [14–16]. This study made conducting leathers using an in situ PEDOT:PSS and dimethyl sulfoxide (DMSO) solution on retanning stages during leather processing stages to enhance the conductivity of leather and comparison with control and treated conductive polymer during finishing stages was done. However, this approach does not produce a three-dimensional fibrous network across the cross-section of leather and does not give uniform conductivity across the surface. Furthermore, it lost its conductivity within a month [2, 17]. All the strategies for introducing electrical conductivity in leather that has been documented involve the retanning process to create a three-dimensional fibrous network throughout the cross-section of the leather. The current research was aimed at developing a simple approach for treating conducting and colored leathers with the conductive polymer during the retanning phases to prepare them for smart or advanced product applications. During the retanning leather manufacturing phases, goatskin was chosen as the substrate and treated with PEDOT:PSS with DMSO as boosts of PEDOT:PSS [18]. The ability to provide both functions of conductivity and color in a single chemical treatment saves time and money.

2. Materials and Methods

Goat wet-blue skin samples were collected from Bahir Dar Tannery, Ethiopia. Sodium formate, ammonia bicarbonate, soda ash, NOVALTAN PF, SINCAL MS, Coriacide blacks NGFloroAl77, Thiosulphate 6146, DMSO, Ombarallon WB. Ombaralons, and formic acid, which were purchased from Zimmer Schwarz Leather Chemicals Company. PEDOT:PSS aqueous dispersion, Clevis PH1000, with a concentration of 1.3 wt%, and PEDOT-to-PSS ratio of 1 : 2.5 was bought from Heraeus, Germany.

2.1. Control and Experimental Treatment Conductivity Polymer during the Retanning Process. The sample wet-blue skins were selected randomly in Bahir Dar tannery. The selected goat wet-blue was wet-back and neutralized and treatments such as dyeing, retanning, and fixation were conducted. To enhance the conductivity, leather was treated with a conductive polymer, and the treatment of conductive polymer PEDOT:PSS with leather was conducted during the retanning and leather finishing stages, each method is described in Figure 1.

2.2. Optimization Recipes for Conductivity PEDOT:PSS Polymer Leather. Processing of conductive leather was conducted by modifying retanning recipes. Processing of the conductivity of glove leather is like conventional leather

Table 1: PEDOT:PSS treatment during retanning with basic dyes stuffs.

Process	Experimental recipe (%)	Conventional recipe (%)
Wetting back	Water (600)	Water (600)
Neutralizing agent	Soda ash (2)	Soda ash (2)
	Novaltan PF (1.5)	Novaltan (1.5)
Washing	Water (500)	Water (500)
Dyeing	Water (500)	Water (500)
	Sinical MS (2)	Sinical MS (2)
	Red dye (13)	Red dyes NG (13)
	Novaltan PF (2)	Novaltan PF (2)
Retanning	Water (250)	Water (250)
	Floro AL 776 (5)	Floro AL776 (5)
	Synthol FL327 (4)	Synthol FL327 (4)
	PEDOT:PSS (0.5)	
Fat-liquoring	Ombarallon WB (4)	Ombarallon WB (4)
	Ombarallon S (4)	Ombarallon WB (4)
Fixation	Formic acid (2)	Formic acid (2)
	Dilution water (5)	Dilution water (5)

processing. It requires tanning and post-tanning, including the addition of color and leather characteristics but modification uses of the chemical during post-tanning, would be conducted. To obtain conductive glove leather the following stages were performed.

2.2.1. Wet-Backing. The collected wet-blue skins were washed with 2% soda ash and 1.5% novaltan PF aqueous solution to prepare for neutralizing processes [19].

2.2.2. Neutralization. One kilogram of goat wet-blue leather was washed in pained water at 32°C in the drum for 15–20 min and 1 L of 1.50% sodium formate and 2.0% sodium bicarbonate were dissolved in 40°C water; half of this solution was added to the drum and run for 20 min; the remaining portion was then added and the drum was run for other minutes and final pH was checked by using bromophenol blue [19].

2.2.3. Retanning by Treating PEDOT:PSS. One kilogram of crust goat skin was taken and washed with 200% water and 1.0% of soda ash to wetback crust for 10 min with cold water. Thirteen percent coriacide black NG black dyestuff were added to drum with 200% of water run for 45 min. To make leather softer 2% sincal MS were used as leveling and Synthol FL327 fat liquor and 50% hot water with 50°C run drums for 1 hr. Finally, PEDOT:PSS polymer was added to the leather substrate by using DMSO as an enhancement, DMSO dispersed in poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) for 10 min before being added to the drum. A solution that contains both PEDOT:PSS and DMSO was added to the drum during the retanning processes after the dyeing process. The effective concentration of conductive polymer is optimized by experiment. Variations in monomer, dopant, oxidant concentrations, and dispersion were used to identify the best treatment conditions for maximal conductivity. The concentrations of PEDOT:PSS (0.05, 0.10, 0.15, 0.20, 0.40, and 0.50 M) and DMSO (0.05, 0.10, 0.15, 0.20, 0.40, and 0.50 M) were varied in the experiments. Before mixing the solutions, the molar concentrations of PEDOT:PSS and DMSO were estimated separately. The treatment lasted 2 hr in a 50°C bath. Finally, formic acid was used to fix the chemical, and the leather was cleaned four times with distilled water before being dried at room temperature for 12 hr before the resistance was measured. Table 1 shows that the application PEDOT:PSS treatment during retanning with basic dyes stuffs.

PEDOT:PSS was pretreated with 0.3% DMSO to remove the hydrophilic PSS toward the surface. This enhances the conductivity of PEDOT. In another experiment, coriacide black was used in the powder form as a dyeing agent, and differences were observed.

2.3. Application of PEDOT:PSS on Resin Finishing. In this method, the prepared solution takes 30 min before being applied to the leather to make a good soluble. After the solution was applied to leather by using a hand spray machine, drying was conducted for 30 min, and the treated leather was passed through a heating machine at 80°C to fix chemicals on leather. In the experiment, two lacquers were applied to leather. In experiment III, the prepared solution was taken for 24 hr to apply on leather to make good dispersion between each other. After the solution was applied to leather by using a hand spray machine drying was conducted for 30 min and treated leather was passed through a heating machine to fix chemicals on the leather (Table 2).

2.4. Characterization of Conductivity Leather. Equations (1) and (2) were used to calculate the electrical conductivity of conductive polymer-treated leathers by measuring resistance using a two-probe approach. The resistance (R) between the two ends of the square specimen was measured and the following equation has been used to estimate the surface resistance.

Table 2: Applications of PEDOT:PSS on resin finishing.

Chemical	Experiment I	Experiment II	Experiment III
Cationic filler	20	20	20
Binder	150	150	150
Pigment	40	40	40
DMSO	5	5	5
PEDOT:PSS	40	50	40
Water	500	500	500
Lacquer	–	1 : 1	1 : 1
DMSO	–	–	5 after 1 day

$$R_S = \frac{RW}{L}, \quad (1)$$

where R_S is surface resistivity, R is the resistance of the sample, W is the width of the sample, and L is the length between electrodes conductivity (S/cm) [20].

$$\text{Conductivity (S/cm)} = \frac{1}{(\text{surface resistivity} \times \text{thickness of leather in cm})}. \quad (2)$$

2.5. Determination of Tensile Strength Conductive Leather. Tensile strength, temporary and permanent elongation at specific loads, modulus, and elongation at break of conducive leather are all determined using ISO 3376:2011/ASTM D2209-00(2015) to prepare the sample pieces. For conducive leather test pieces, set the jaws of the tensile strength machine 20 mm apart. Clamp the test piece in the jaws so that the jaws' edges are parallel to the lines when clamped, and the test piece's grain surface is in one plane. Run the machine until the test component breaks, then set the breaking load to the highest load reached.

2.6. Method for Measurement of Tear Strength. The load in kilograms was required tearing a conductive glove leather having a thickness of 1 cm. Measure the thickness of conditioned test pieces according to ISO 3376:2011/ASTM D2209-00(2015). Run the machine until the test piece is completely shredded. As for the tear strength, note the peak load reached during tearing.

2.7. Method of Test Colorfastness. The method specifies rubbing one side of the glove leather to be evaluated with pieces of reference wool felt under a given pressure for a specified number of forwarding and backward motions, then evaluating the change in color of the pieces of felt and the leather with the gray scales test according to ISO 1164:1993.

2.8. Organoleptic Properties of Control and Experimental Sample Leathers. Organoleptic was conducted by nine experienced leather technologists and the average was taken. The conventionally and experimentally treated leather had the same result on grain pattern, uniformity of color, and fullness but difference between feel and general appearance, experimentally treated leather shows a better result on feel and general appearance.

2.9. Characterization of Control and Experimental Sample Leathers by FTIR. Treated leather with conductive polymer and untreated leathers were further characterized using Fourier-transform infrared (FTIR) spectroscopic measurements using a PerkinElmer FTIR spectrometer (USA) in KBr medium in the region of 4,000–500 cm^{-1}.

2.10. Characterization of Conductive Leather by SEM. Scanning electron microphotographs of chrome- and direct-tanned crust were compared to analyze the influence of tanning on the structural properties of the leathers produced. Fresh stainless-steel blades were used to cut the 5×2 mm samples from the crust leathers. Using an adhesive, the samples were placed vertically and horizontally on aluminum stubs. After that, an Edwards E-306 sputter coater was used. An FEI-Quanta 200 scanning electron microscope was used to place the stubs in the specimen chamber. The stage's stubs tilted, rotated, and shifted to any desired position and orientation. The cross-section micrographs were created by running the microscope at 5 kV.

3. Results

This section will highlight the results of the experiments and divided into various sections as follows.

3.1. Neutralization. The chrome-tanned leather can have free acid, protein-bound acid, and acid present inside the coordination sphere of the chrome complex and fixed up with the chrome complex by primary valence. During neutralization, free acid and protein-bound acid are efficiently removed either by repeating the next steps like dyeing-tanning or fat-liquoring were not properly performed. After neutralization, our leather by sodium formate and sodium bicarbonate is not aged for a time otherwise the free acid out from the middle layer due to hydrolysis of the chrome complexes will after pH. At the ends of tanning leather is at a pH was around 4.0 when leather is treated by chrome tanning the pH was decreased due to olation since protons were released by hydroxy bridges of chrome complexes. To adjust the isoelectric points of skin, the pH value was raised by using mild alkaline salts such as sodium formate removing all acid even the acid groups present inside the coordination spheres of chrome complexes, which are not desirable.

3.2. Comparison of Treating PEDOT:PSS with DMSO on Retanning vs. Resin Finishing. Retanning was a process conducted post-tanning after the tanning process, but finishing was the final stage of leather processing. During retanning processing, chemical reactions were conducted between chemical and fiber collagen, but during finishing, the most mechanical reaction was conducted rather than a chemical reaction. Chemical interaction between fiber and applied on leather less and mostly coating happened. The polyion complex disperses in water as a colloidal gel particle with a diameter of 10 nm where hydrophobic PEDOT:PSS molecules aggregate to form physical cross-links of the PSS chain

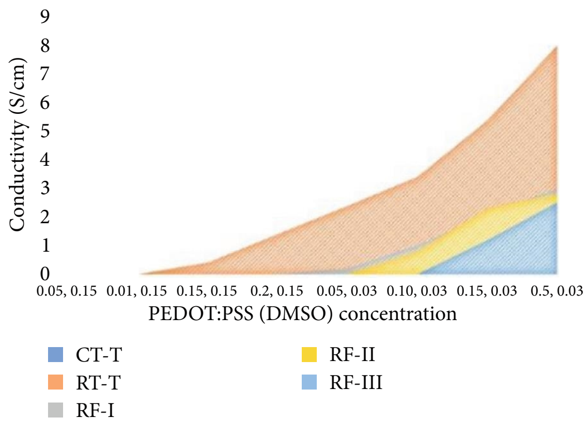


Figure 2: Comparison of electrical conductivity characteristics of conventional and treated leather.

colloidal gel particles can be shaped into collagen fibers. Under this study, leather treated under retanning showed good conductivity rather than finishing processes. In this study, leather treated under finishing operations and untreated did not show a significant difference. The interaction between fiber and PEDOT:PSS with a DMSO enhancement reaction by decreasing the interchain interaction between conductive PEDOT-rich grains and the insulating PSS-rich shell and it can dissolve some unassociated PSS phase from the film surface during the retanning process after leather acid dyeing induced remarkable results on black acid dyes more than red basic dyes. This implies the conductivity of PEDOT:PSS with the enhancement of DMSO had good reactivity and stability with acids and black dyes. Conductivity was unstable during the application of PEDOT:PSS with DMSO during finishing due to a lack of reactivity between conductivity polymer and leather fiber.

3.3. Electrical Conductivity Characteristics. Initial tests were conducted to find the optimum doping agent, dispersion time, and temperature for producing conducting leather, but the concentration of PEDOT:PSS has a significant impact. Figure 2 shows that the increased concentration of PEDOT:PSS increases the conductivity of leather.

When retanning goat leather using a leather drum, the conductive qualities were created by combining 0.3 M DMSO with 0.5 M poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS). The final leather had a conductivity of 8.0 S/cm, however, in comparison to other research, conductive leather finished by coating had a conductivity of 7.2 S/cm [20]. The comparison results show that the technique, raw skin species, and polymer employed are all distinct in addition to the finishing steps. By using in situ double polymerization at 5°C of 0.3 M pyrroles, 10% anthraquinone-2-sulfonic acid sodium salt monohydrate as the dopant, and 2.67 M ferric chlorides as the oxidant on leather finishing stages, the author created gloves made of sheepskin leather that could operate smartphones, tablets, and iPods.

3.4. Physical and Mechanical Properties of Experimentally Treated Leather vs. Controlled Leather. The physical analysis was conducted after leather was treated under conventional and experimental pass out under the same all-leather processing stages and finally, the leather character was compared, as shown in Figure 3, the physical strength properties of the leather. The leathers obtained from all the experiments RT-T show comparable tensile and tear strength values relative to conventionally treated leather. The strength property is the evidence that the leather produced in the experiment does not show any alteration in the leather property.

3.5. Rub Fastness Characteristics of Control Leather vs. Experimental Leather. Although the PEDOT:PSS-treated leathers resulted in generated black conducting leathers without employing toxic dyes, the color fastness of the treated leathers analyzed. As shown in Figure 4, dry as well as wet rubbing fastness for leathers treated during retanning was more than that of the standard requirement. This may be due to the higher absorption of reactants into leather owing to a long dispersion time, which reduces excess monomer deposition on the surface.

3.6. Organoleptic Properties. Organoleptic was conducted by nine experienced leather technologists and the average value was taken. The conventionally treated leather and experimentally treated leather the result showed little modification properties on RT-T and to some extent like conventional leather, but RF-I shows better fullness (Figure 5).

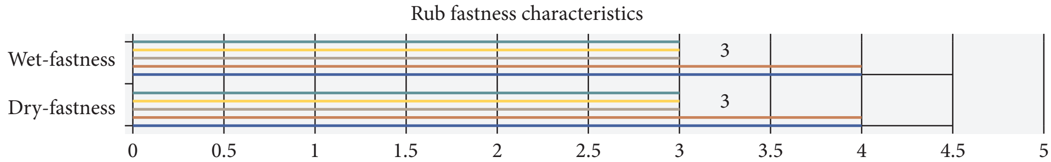
3.7. Infrared Spectroscopy Observations. Figure 6 shows the FTIR spectra of control leather and experimental leathers treated with PEDOT:PSS. The FTIR spectra of control leather possess characteristic peaks. RT-T leather formed a single bond on 3,315.65 cm^{-1} with 0.73 peaks intensifying and forming singles bonds (–OH) and (NH_2) on 2,921.39 cm^{-1} alkyl (–CH_2), 1,663.70 forms carboxyl group and amide group. The FTIR spectra of control leather possess characteristic peaks corresponding to amide I (1,700–1,600 cm^{-1}), amide II (1,600–1,500 cm^{-1}), and amide III (1,300–1,200 cm^{-1}) bands. Further, it also shows an additional peak at 1,458 cm^{-1} corresponding to aliphatic side chain groups of amino acids present in collagen. These characteristic peaks of collagen were not significantly altering PEDOT:PSS (DMSO).

3.8. Surface Morphology using SEM. Scanning electron microscopic images showing the surface of untreated and treated leathers are shown in Figure 7. Insets shown in the SEM images are a magnified view of the selected portion of the images. Untreated leather (Figure 7(a)) shows a clear, smooth surface and hair pores free from any foreign particles. On the other hand (Figure 7(b)), the micrographs of RT-T-treated leathers show polymeric particles of PEDOT:PSS with DMSO on the surface, which can be attributed to the conducting nature of treated leathers but affinity between fibers was visible. Figure 7(c) shows that the area of leather is highly deposited in the polymer, which does not interact with collagen fiber rather than precipitates on the surface. Hence, reaction parameters such as soaking time, dispersion time, reactant concentration, and dispersion were influenced by reactivity and affinity PEDOT:PSS to collagen fibers.

	Tensile strength (kg/mm^2)	Tear strength (N)	Elongation (%)	
CV-T	36.1	37	56	
RT-T	38.21	39	59.2	
RF-1	37.5	35.4	57	
RF-2	35.8	37.2	58	
RF-3	38.4	36.4	57.2	

CV-T
RT-T
RF-1
RF-2
RF-3
Linear (CV-T)

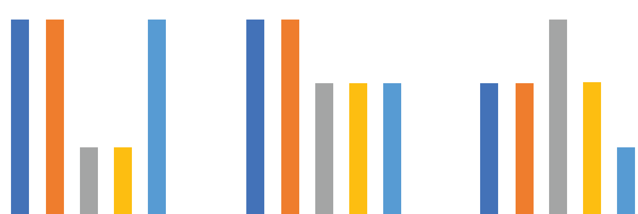
Figure 3: Physical and mechanical properties of experimental conductive leather vs. control leather.



	Dry-fastness	Wet-fastness
RF-III	3	3
RF-II	3	3
RF-I	3	3
RT-T	4	4
CT-T	4	4

RF-III
RF-II
RF-I
RT-T
CT-T

Figure 4: Comparison of rub fastness properties of conventional and treated leather.



	Softness	Roundness	Fullness
CV-T	8	8	7.5
RF-T	8	8	7.5
RF-I	7	7.5	8
RF-II	7	7.5	7.5
RF-III	8	7.5	7

CV-T
RF-T
RF-I
RF-II
RF-III

Figure 5: Compassion organoleptic properties of conventional and experimental treated leather.

3.9. Application Study. The ability of conductive goat leather-treated poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) and DMSO was demonstrated. During the demonstration, both control light blue goat leather and treated black leather were used for operating smart touch-screen mobile phone devices (see https://www.youtube.com/shorts/zt6UDrRajeU). As can be seen, the control light blue leather could not operate the device well, but black leather

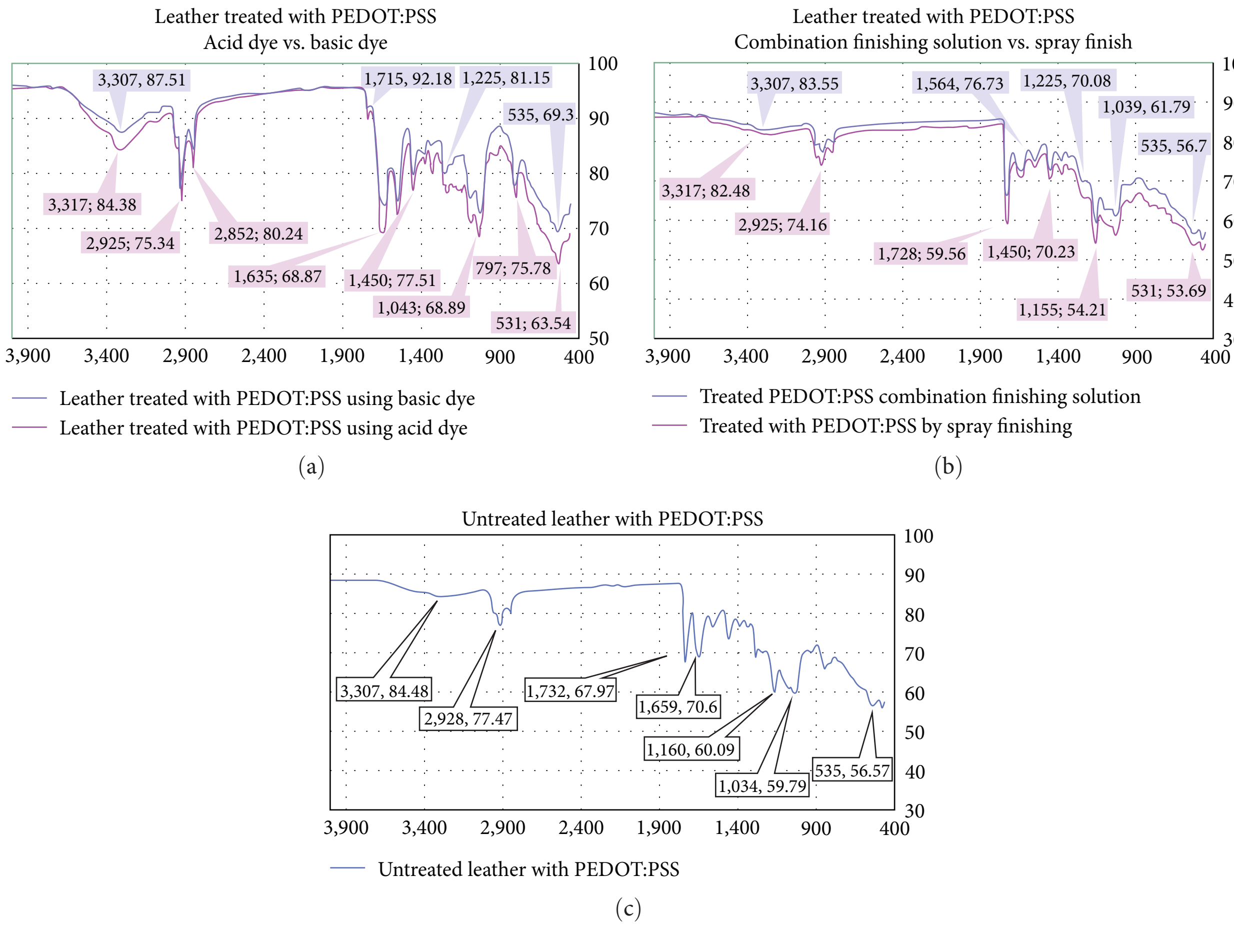


Figure 6: (a) Leather treated with acid vs. basic dyes; (b) leather treated with PEDOT:PSS combination finishing solution vs. spray finish; and (c) untreated leather with PEDOT:PSS.

treated during retanning stages in this study can be shown to potentially be used for smart or advanced product applications.

4. Conclusions

A straightforward process to prepare and conduct leathers through conductive PEDOT:PSS polymer with DMSO as enhancements shows excellent results on black dyestuff during the retanning process than another stage of the leather process. 0.5 M of PEDOT:PSS, a dopant concentration of 5 wt% of DMSO enhancing agents were used to make highly conducting leathers with a conductivity value of 8.00 S/cm. When PEDOT:PSS was added to the solution, the tanning materials show excellent conductive properties because they contain polar groups. As these materials are not compatible with the active groups in collagen fibers of leather while maintaining their polarity, they show electrical conductivity. Leather has a higher conductivity when it has more polar groups. When cation retention agents were used with PEDOT, the leather will show positive electricity; PSS and DMSO as enhancing agents, on the other hand, will show negative electricity. The retention agents were mixed with the opposing electric charge, and the electric charge of the two materials was entirely neutralized by the polar group of leather, resulting in lesser conductive qualities of the leather. Cation and anion fat-liquoring agents all have electric properties. When leather was treated it acquires an electric charge or the polar group was neutralized, resulting in a change in conductive properties. The fat-liquoring agent, as well as PEDOT:PSS exhibits evacuation and lubricating on the fiber of leather, exposing the polar group in the leather, resulting in conductive qualities in the crust leather. Using high amount fat-liquoring agent will parcel the surface of collagen fibers, the greater the degree of separation between the fibers, the greater degree of isolation between the fibers, the fewer polar groups were exposed, resulting in reduced conductive qualities of leather. The more pressure applied, the thinner the leather becomes, and the leather fibers get tighter. As a result, the polar group of collagen fibers was brought together more intensely than before, and the polar group's conductive qualities improve. With the increase of relative humidity and the rise of moisture content in the leather, it can make polarity materials and polar groups, which were not charged before, ionized, and display electrical properties. The stronger the ionization degree, thus the conductive properties are stronger. The result also showed that leather treated during the retanning stage with acid dyes was highly conductive than resin finishing and conventionally treated leather, under the finishing stage.

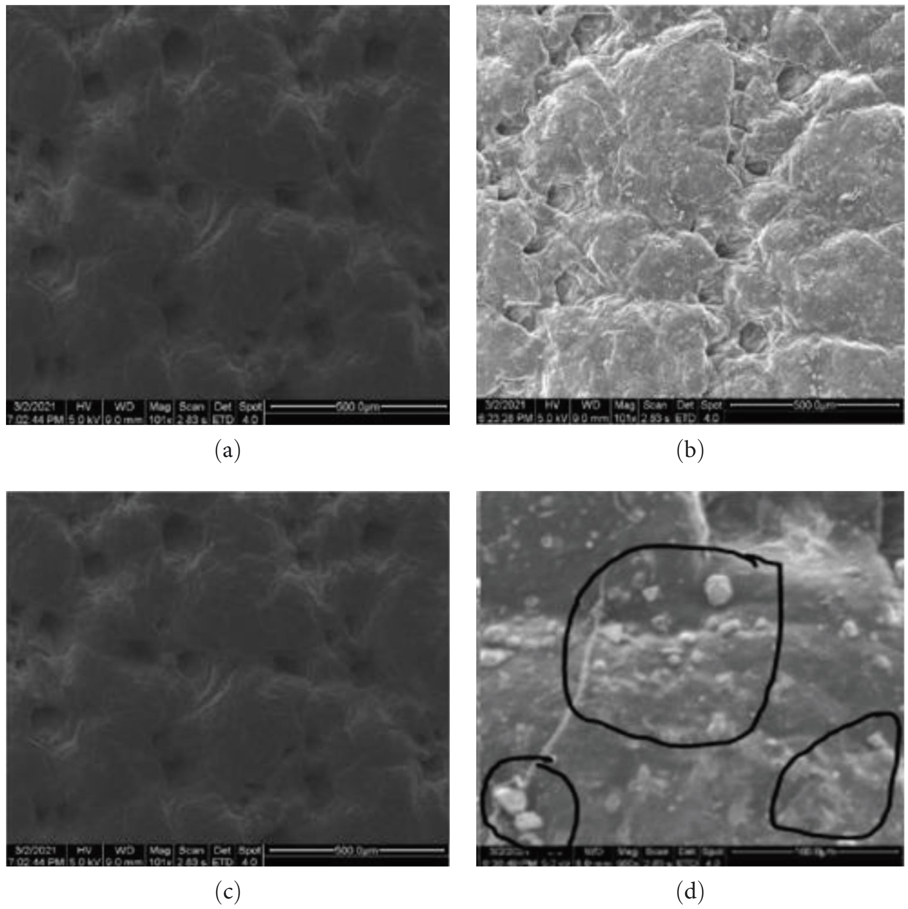


Figure 7: SEM micrographs of the grain surface: (a) CV.T vs. (b) RT-T and (c) CV.T vs. (d) RF-I.

Data Availability

The data collected and analyzed during this study are included in the paper and can be accessed more from the authors through a rational request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Authors' Contributions

Conceptualization: Z. T. Kebede, M. G. Tadesse, and D. A. Mengistie; methodology: Z. T. Kebede and M. G. Tadesse; validation: Z. T. Kebede, M. G. Tadesse, and D. A. Mengistie; formal analysis: Z. T. Kebede and M. G. Tadesse; investigation: Z. T. Kebede and M. G. Tadesse; writing—original draft preparation: Z. T. Kebede and T. E. Chane; writing—review and editing: Z. T. Kebede and M. G. Tadesse; all authors have read and agreed to the published version of the manuscript.

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