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Leather Finishing Using Nanomaterials for Advanced Properties

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Abstract

The aim of this presentation is to highlight how various nanomaterials and nanocomposites can impart multifunctional properties to leather surface. Key findings on the use of nano titanium dioxide decorated with nitrogen, silver, silica, copper oxides, or graphene oxide, as well as nano zinc oxide or multiwall carbon nanotubes are presented based on prior experience in nanoparticle application in leather finishing. The multifunctional properties of leather surfaces coated with these nanomaterialsincluding antibacterial, antifungal, and selfcleaning effects against organic soils and volatile organics, electrical conductivity, and UV light shielding properties are presented.

The last achievements in using silica and magnetite nanoparticles, with lower toxicological impact, for medical footwear components will be shown. The processing of leathers by integration the silica, magnetite with or without natural extracts (essential oils) in classical finishing technologies will be approached. The antimicrobial, self-cleaning and leather surface fastness resistances (abrasion, rubbing, water drop tests) will be critical discussed. SEM/EDX analyses, biological resistance against fungi, gram-positive and gram-negative bacteria, photodegradation of organic stains under visible light exposure as well as the cytotoxicity concentrations and

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anti-inflammatory potential of the leached solutions will be presented. The main conclusions were that the leathers coated with silica or magnetite nanoparticles have the potential to protect the feet of diabetic patients inside medical footwear without harmful impact on skin and environment.

Keywords: nanomaterials, leather coating, smart properties, surface properties

1. Introduction

Leather surfaces can be functionalized for smart properties development by using nanomaterials or nanocomposites as alternatives to organic, volatile chemical products. The improved durability and antimicrobial protection selfgenerated by visible light exposure made leather more ecological and protective against dirt and microbial spreading, but also offering other properties like fastness resistances [1, 2], electric conductivity for touch screen applications or UV shielding [3]. Leather durability through self-cleaning properties represents a principle stipulated by ecological labels and means that the leather articles will have a longer life without the need for cleaning and the use of organic solvents. Enhanced antimicrobial properties can bring a safer life and less economic loss due to disease spread and the use of nondurable antimicrobials. Self-responsive surfaces under UV light exposure were made through the use of nanomaterials based on titanium dioxide [4], meanwhile, the last

generation of nanocomposites showed enhanced reactivity under Vis light. The use of nanomaterials based on silica in combination with silver [5] or natural active substances [6] showed the potential to act synergically against viruses or other resistant pathogens. The performances of nano silica and nano magnetite deposited on leather surface alone or in combination with natural products like essential oils are presented as potential pathway in antimicrobial footwear design.

2. Methodology

Leathers for footwear components: pig skins (Fig.1 a, b), goatskins (Fig.1 d), bovine leathers ((Fig.1 d) were processed in Leather Department to the crust stage. Silica nanoparticles with 15-20 nm average size and magnetite with 50 nm average size were purchased from chemical companies as well as chemical auxiliaries for leather finishing. Leather coating was performed by spraying (Fig 1c) in a similar manner as in industrial conditions, by alternating the base coat with water dispersed solutions of nanoparticles with and without essential oil (thyme). The top coat was a classical water emulsion of nitrocellulose lacquer. The antimicrobial resistances of leather surfaces were assessed following adapted methods according to ISO 19574:2021 and ISO 16187:2025. The self-cleaning test was performed by simulating the organic stain with methylene blue and measuring the initial color and the color modification in time, after exposure to UV and Vis light with the photocolorimeter Ci7600 (400) Benchtop Spectro equipped with Color iQC Basic Perpetual License. The influence of nanoparticles on leather surface fastness was assessed according to standards in force. In order to estimate the impact of new coatings on human skin and on the environment by leaching on the surface, the cytotoxicity concentrations determination and anti-inflammatory tests were performed following in vitro cytotoxicity testing by quantitative MTT method and anti-inflammatory activity determination by dosing specific cytokines with ELISA kits. The levigates were prepared according to SR EN ISO 4098:2018. The study of water drop contact angle in dynamic conditions

was performed using a VGA Optima XE device in view of leather behavior identification under UV and visible light exposure, induced by nanoparticles. The morphology and elemental composition of leather surface were done using FEI Quanta 200 SEM equipped with EDX.

(a) (b)

(c) (d)

Fig.1. Leather samples coated with NPs: pig skins

coated with (a); magnetite NPs (b) silica NPs; (c) spray coating of leather footwear components; (d) bovine leathers and goat skins coated with magnetite

and silica for footwear manufacturing

3. Result & Discussion

The antimicrobial tests performed on leather coated with silica or magnetite nanoparticles with or without thyme essential oil (EO) showed that the resistances against Candida albicans, Aspergillus niger, Escherichia coli and Staphylococcus aureus were improved as compared to control samples (Fig.2) confirming the potential of nanoparticles to generate oxygen reactive species (ROS) under visible light. As compared to other nanoparticles, silica and magnetite are considered more environmentally friendly due to the wider distribution of silicon and iron in nature. The embedded nanoparticles in film forming polymers represent also a less harmful use of nanoparticles.

(a) (b)

(c) (d)

Fig.2. Antimicrobial resistances of leather surfaces coated with NPs against: (a) Candida albicans;

(b) Aspergillus niger; (c) Escherichia coli and (d) Staphylococcus aureus.

The generation of ROS allows to decompose not only

biological load on the leather surface but also the organic

dirt. In this regard, self-cleaning tests against methylene

blue allowed to demonstrate that the leather surface has

new advanced, smart properties. In Figure 3 can be seen

that the organic stain was 30-50% decomposed as

compared to 10-13% for the control samples and the

presence of EO decreased the self-cleaning efficiency.

The generation of reactive oxygen species under visible light showed to be more active as compared to UV light which can be an advantage for the efficiency of leather functionality (antimicrobial, self-cleaning) in daily life. Figures 4 a and b present the water drop dynamic contact angle in simulated condition of exposure to visible and UV light, where can be seen a more hydrophilic surface under visible light exposure, in time, as compared to UV light exposure.

(a) (b)

Fig.4. Water drops contact angle on leather surface exposed to UV and Vis light

The leather morphology analyses performed by scanning electron microscopy showed a uniform distribution of nanoparticles with nano agglomerated depositions and the

Fig.5. SEM/EDX images of leather surface coated with Fe_3O_4 NPs and SiO_2 NPs

The leached solutions in the case of goat skins and bovine leathers coated with NPs showed slightly higher

Table 1. Soluble substances of leathers coated with nanoparticles

Name of the test Technical characteristic	BM	BP	NM	NP
Determination of the content of water-soluble substances				
Total soluble substances, %	3.28			
Soluble mineral substances, %	0.60			
(SR EN ISO 4098 :2018)				
Soluble organic substances, %	2.68			

elemental mapping generated by Energy Dispersive Xray Spectroscopy allowed identification Fe and Si on leather surface (Fig.4).

concentrations in total soluble substances and mineral and organic components (Table 1).

Samples/Values	Goat Skin		Bovine Leather	
	Control Sample		Control Sample	
	3.96			
	0.63			
	1.04			
	0.10			
	0.35			
	0.68			
	0.53			
	0.69			
	3.28			

The evaluation of cytotoxicity concentrations (Fig.6) revealed no cytotoxic effect after 24 hours of contact and cytotoxic effect for 25% concentration after 48 hours of contact for nano silica levigates. The cytotoxic effect of control sample at 50% concentration suggests

Fig.6. Viability of NCTC fibroblasts cultured in the presence of BM (goat skin-control), BP (goat skin coated with SiO_2 NP), NM

(Bovine leather -Control) and NP (Bovine leather coated with F3Os NP) samples, assessed by MTT assay at 24 and 48 hours.

The control (untreated cells) to which all samples were reported was considered to have a viability of 100%.

The tests for anti-inflammatory effects of leather levigates showed that the goat skins coated with silica nanoparticles have anti-inflammatory effect (Fig.7 a, b, BPP-C sample) as compared to control sample for

Fig.7. (a) Measurement of TNF-α cytokine secreted by THP-1 cells stimulated with LPS and treated with

different concentrations of the samples of interest, by ELISA method; (b) Measurement of IL-1β cytokines secreted by THP-1 cells stimulated with LPS and treated with different

concentrations of levigates, by ELISA method. (BM-P-pig skin control; BP-P-pig skin coated with SiO2 NPs; BMP-C-goat skin control; BPP-C-goat skin

coated with SiO2 NPs)

that there are other classical components with this

effect. The presence of magnetite on leather surface

showed less cytotoxic effect, the concentration of 50%

being as much cytotoxic for control and sample.

both inflammatory markers, meanwhile the pig skins coated with nano silica (BP-P) presented similar behavior as compared to control sample (BM-P).

The main surface fastness resistances exemplified in Table 2 for nano silica coated skins show insignificant differences compared to the control sample.

Table 2. Fastness resistances of leather surfaces coated with SiO2NPs as compared to control sample

Name of the test

Technical characteristic

UM

Finished leather -

Physical and mechanical tests for

Determination of

Cycles

25600 6400 25600 6400

abrasion resistance

soiling - Part 1: Rubbing method

Without

Remarks

modifi-

(Martindale)

cations

after

30 min

Marks

Leather. Resistance of

Determination of

dye to water drops

resistance to

water drops

after

Determination of

Finished leathers.

dye resistance to

Dye resistance

rubbing (50 cycles) Leather-

tests. Dye resistance to back-

Marks

Felt

and-forth rubbing

4. Conclusion

Leather coated with silica or magnetite nanoparticles with or without essential oils showed antimicrobial, self-cleaning and standard fastness resistance of surface and potential to be used in medical footwear manufacturing. The cytotoxicity and anti-inflammatory tests performed on soluble substances extracted from leather coated with nanoparticles suggest the innocuity

Sample/Values

The method standard

Pig skin coated with

Pig skin-Control

SiO2NPs

BM-P

BP-P

Dry Wet Dry Wet

SR EN ISO

260821:2019

Without

Without

Without

modifi-

modifi-

modifi-

cations

cations

cations

5

5

after 30

The drop

The drop

min

was absorbed without a

was not absorbed (remained

SR EN ISO

15700-

without a

stain

2000

stain)

5

5

STAS 8259/

after

The drop

The drop

was absorbed without a

was absorbed without a

16 h

16 h

3-82

stain

stain

Dry Wet Dry Wet

SR EN ISO

11640-

5-5

5-5

5-5

5-4/5

2019

of the leather coatings for human skin and environment. The application of new nanomaterials was performed by classical technology of leather finishing and opens the pathway for larger experiments and wearability test in view of validation for medical use.

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