BIOACTIVE TEXTILES OBTAINED BY APPLYING CINNAMON ESSENTIAL OIL-BASED EMULSIONS

LAURA CHIRILA¹, ALINA POPESCU¹, SABINA OLARU¹, ANGELA DANILA²,
RODICA ROXANA CONSTANTINESCU³, ANDREEA TIGAU¹

¹National Research & Development Institute for Textiles and Leather, 16 Lucretiu Patrascanu Street, 030508, Sector 3, Bucharest, Romania, laura.chirila@incdtp.ro

²“Gheorghe Asachi” Technical University of Iasi, Faculty of Industrial Design and Business Management, 29 Prof. Dr. Doc. Dimitrie Mangeron Blvd., 700050, Iasi, Romania

³INC DTP - Division: Leather and Footwear Research Institute (ICPI) Division, 93 Ion Minulescu Street, 031215, Bucharest, Romania

In order to obtain biomaterials with potential use in the treatment of inflammatory skin conditions, this experimental study approached the immobilization on knitted fabric, made of 100% organic cotton, of oil-in-water emulsions type, based on xanthan-propolis-cinnamon essential oil, in certain formulations. For this purpose, seven experimental variants of emulsions were prepared and characterized, by specific methods, from physical-chemical and stability point of view. The functionalized textile materials were also characterized from morphological and antibacterial activity view point. The stability index, determined after 10 days, indicated that the emulsions are stable, without showing the presence of one of the flocculation, creaming / sedimentation, coalescence or Ostwald ripening phenomena. The lowest value of the turbidity was obtained for the experimental variant containing the lowest amount of essential oil and propolis. For the same variant, the highest value of viscosity was obtained, in which 0,363 mL water /mL emulsion and 0,5 mL xanthan/mL emulsion volume fraction was used. The textile materials treated with the synthesized emulsions based on xanthan-propolis-cinnamon essential oil shows antibacterial effect against *S. aureus* and *E. coli* test strains.

Keywords: bioactive textiles, xanthan gum, cinnamon essential oil.

INTRODUCTION

Textiles are an excellent carrier of different bioactive compounds through contact with the skin and have found applications for medical, hygienic, and health purposes (Alonso *et al.*, 2013). Medical textiles are the products and constructions used for medical and biological applications such as first aid, clinical and hygienic purposes. With the growing of public awareness of potential health risks, antimicrobial properties have attracted increasing attention among practitioners and have been successfully imparted to textiles to improve the resilience against microorganisms (Dastjerdi and Montazer, 2010; Yuan and Cranston, 2008; Windler *et al.*, 2013). Currently, antimicrobials have been playing an increasingly important role in addressing textile hygiene in clinical and sensitive environments. The development of antibacterial textiles has received extensive attention and applications in several skin disorders. This proposal was developed on the basis of the fact that a majority of skin disorders patients suffer from recurrent bacterial infections, therefore, antimicrobial textiles can be desirable alternative therapy to treat several skin inflammations by killing bacterial colonization. In order to obtain the textile materials with potential for use in the treatment of inflammatory skin conditions, this study approached the immobilization on knitted fabric, made of 100% organic cotton, of bioactive polymeric systems in certain formulations, based on xanthan-propolis-cinnamon essential oil.

EXPERIMENTAL PART

Materials

Xanthan gum (Mayam, Romania) was used as an embedding agent of bioactive agents. Tween 80 (Sigma Aldrich, Germany) was used as a surfactant with emulsifying role and glycerol (Honeywell, USA) it has been used as a solubilizing agent with wetting role. Cinnamon leaf essential oil (Mayam, Romania) and propolis (tincture of 70% ethanol solution) (Larix SA, Romania) were used as bioactive agents. For preparation of bioactive systems the distilled water has been used. Bleached 100% knitted organic cotton was used for the functionalization processes.

Emulsion Preparation Methodology

For the achievement of the bioactive systems, initially stock solutions of 1% xanthan gum and 30% Tween 80 solution were prepared. Further, over the previously prepared xanthan solution, glycerol was added dropwise and the system was maintained under magnetic stirring for 10 minutes. After that, Tween 80 emulsifier and distilled water separately were added under vigorous magnetic stirring. Further, after complete homogenization of the solution, the propolis and cinnamon essential oil were separately added, maintaining the magnetic stirring for 10 minutes each stage. Thus, 7 variants of emulsions have been developed that differ by: i) the nature and concentration of the embedding agent, ii) the concentration of the selected bioactive compounds and iii) the concentration of the dispersion medium (water). The selected experimental variants are presented in Table 1.

Table 1. The concentration of constituents of xanthan gum-propolis-cinnamon essential oil polymeric system

Code	Composition of the emulsions (reported of 30 mL emulsion)					
	Xanthan (mL)	Tween 80 (mL)	Glycerol (mL)	Water (mL)	Essential oil (mL)	Propolis (mL)
R1	9	0.67	3	16	0.67	0.67
R2	12	0.67	3	13	0.67	0.67
R3	15	0.67	3	10	0.67	0.67
R4	18	0.67	3	7	0.67	0.67
R5	15	0.67	3	10.9	0.22	0.22
R6	15	0.67	3	10.45	0.44	0.44
R7	15	0.67	3	9.57	0.88	0.88

Immobilization of Bioactive Polymeric Systems on the Textile Materials

The 7 experimental variants were immobilized on the textile materials by the padding method on the laboratory padder (ROACHES, UK). The treated textile materials were then subjected to the drying operation at 50°C for 3 minutes.

Methods

Optical Microscopy

The prepared emulsions were analyzed microscopically using an OLYMPUS BX51 optical microscope (Philippines) equipped with the OLYMPUS UC30 photodigital camera.

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Determination of the Emulsion Stability Index

Immediately after preparation, the emulsions were introduced into test tubes and the evolution was followed for 10 days after their preparation. The stability of the emulsions was evaluated by determining the stability index (ESI) using the relation:

$$ESI (\%) = H - (H_s + H_c) / H \quad (1)$$

where: H – the initial height of the emulsion [cm], H_S - the height of the serous layer [cm]; H_C – the height of the creamy layer [cm].

Turbidity Analysis

In order to determine the turbidity, the transmittance was determined, using a Camspec M501 UV-VIS Spectrophotometer. Turbidity was calculated with the relationship:

$$\tau = 2,303A / L, \quad (2)$$

where: τ – turbidity [cm⁻¹]; A - absorbance; L - path length of cuvette [cm].

Viscosity Determination

The viscosity measurements of the obtained bioactive systems were performed with the DV2T Brookfield AMETEK viscometer. Measurements were performed in triplicate.

Electron Microscopy

Visualization of the morphology of the cotton fiber surfaces treated with the bioactive systems was performed by scanning electron microscopy using the Quanta 200 electron microscope (FEI, The Netherlands).

Assessment of Antibacterial Activity

The antibacterial activity of the treated samples in different variants was qualitatively assessed by Agar diffusion method according with the SR EN ISO 20645:2005 standard - Determination of antibacterial activity-agar diffusion plate test, by using of cultures in liquid medium replicated at 24 hours of ATCC 6538 *Staphylococcus aureus* and ATCC 11229 *Escherichia coli* test strains. The textile specimens (2 cm diameter) are placed on the surface of the nutrient medium and then incubated at 37°C for 24 h. Inhibition zones were calculated using the following formula:

$$H = (D - d) / 2 \quad (3)$$

where: H is the inhibition zone [mm]; D – the total diameter of specimen and inhibition zone [mm]; d– the diameter of specimen [mm].

RESULTS AND DISCUSSION

The images obtained by optical microscopy for the 7 experimental variants are shown in Figure 1. According to microscopic images, the dispersed molecule phase is presented as a compact, dense small globule mass. Normally, the destabilization of the emulsions begins with the drops flocculation of essential oil followed by the phenomenon of coalescence and formation of two distinct phases (separation).

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Figure 1. Optical microscopy of the resulted bioactive systems

Emulsions Stability

The calculated values for the stability index, are shown in Table 2. The values recorded for the stability index over a period of 10 days indicate that they were stable, during this time not showing the presence of one of the flocculation, creaming / sedimentation, coalescence or Ostwald ripening phenomena. Generally, the turbidity of the bioactive polymeric systems is correlated to a great extent with their stability. Analyzing the values obtained for the 7 experimental variants it can be observed that the lowest values of the turbidity were obtained for the R5 variant (1.90) in which the smallest amount of active principle is also present (essential oil and propolis). Turbidity decreases with increasing particle size of emulsion.

Table 2. Emulsions stability

Stability	R1	R2	R3	R4	R5	R6	R7
Turbidity [cm^{-1}]	2.96	3.07	3.57	3.41	1.90	2.27	3.45
Stability index [%]	100	100	100	100	100	100	100

Viscosity

The main viscosity indices recorded for the synthesized emulsions are shown in Table 3.

Table 3. Viscosity indices recorded for the resulted emulsions

Code	Viscosity [cP]	SS [dyne/cm ²]	SR [s ⁻¹]	Speed [RPM]	Temperature [°C]
R1	35,41	65,87	186	200	24,9
R2	45,91	85,40	186	200	25,2
R3	53,08	98,74	186	200	25,3
R4	61,30	114,10	186	200	25,3
R5	64,08	119,16	186	200	25,4
R6	57,75	107,43	186	200	25,3
R7	52,25	97,18	186	200	25,3

From the comparative analysis of the viscosities values it is observed that the highest value was obtained for the experimental variant R5 (64.08 cP) in which in which 0,363 mL water /mL emulsion and 0,5 mL xanthan/mL emulsion volume fraction were used. It can also be observed that the viscosity of the system is not dependent on the volume fraction of active principle (propolis and essential oil), the highest viscosity was obtained when using smaller volume fraction ($\phi = 0.015$ mL oil / mL emulsion). The shear stress (SS) has the highest value in the case of the R5 variant, the variant obtained when using the smallest volume fraction of water.

Electron Microscopy of Textile Materials Functionalized with Bioactive Systems

The images obtained at a magnification of x 8000 for knitted textiles treated with bioactive systems are shown in Figure 1. The resulting micrographs reveal that the surface of the cotton fibers is covered with a thin polymeric layer deposited both on the surface of the fibers and inside the space between the fibers. However, the micrographs cannot provide information about the thickness of the deposited layer and its uniformity.

Figure 2. Electronic images recorded for the knitted fabrics treated with xanthan-propolis-cinnamon essential oil polymeric system

Antibacterial Activity

Images of Petri plates after 24h incubation are shown in Figure 3 and assessment of antibacterial activity is shown in Table 4.

Figure 4. Images of Petri plates showing antibacterial effect after 24 h of incubation

By analyzing the obtained results, it can be concluded that the textile materials treated with synthesized emulsions based on xanthan-propolis-cinnamon essential oil have antibacterial effect against the *S. aureus* test strain, with inhibition zones between 1mm (R5) and 5mm (R2). In the case of *E. coli* strain, the presence of the inhibition zone and the growth of the test strain in the whole culture medium were not observed, being considered a satisfactory effect in terms of antibacterial activity.

Table 4. Evaluation of the antibacterial activity

Code	<i>E. coli</i>		<i>S. aureus</i>	
	Inhibition zone [mm]	Evaluation	Inhibition zone [mm]	Evaluation
R1	0 (*)	Satisfactory effect	0 (*)	Satisfactory effect
R2	0 (*)	Satisfactory effect	5	Satisfactory effect
R3	0 (*)	Satisfactory effect	3.5	Satisfactory effect
R4	0 (*)	Satisfactory effect	3	Satisfactory effect
R5	0 (*)	Satisfactory effect	1	Satisfactory effect
R6	0 (*)	Satisfactory effect	0 (*)	Satisfactory effect
R7	0 (*)	Satisfactory effect	0 (*)	Satisfactory effect
M	0	Unsatisfactory effect	0	Unsatisfactory effect

(*) - no inhibition zone, no multiplication

CONCLUSIONS

Seven experimental variants of bioactive polymer systems synthesized based on xanthan-propolis-cinnamon essential oil were assessed to explore their physico-chemical, biological as well as functional property to understand the possible applications in developing of biomedical textiles. From the cumulation of the obtained data it was found that the prepared emulsions have a high degree of stability and a suitable viscosity to ensure a uniform deposition on the textile materials, on the one hand, and on the other hand to be able to be kept in a single phase until further application on textile materials. Textile materials treated with bioactive polymeric systems have shown antibacterial activity for both gram positive bacteria (*S. aureus*) and gram negative bacteria (*E. coli*) test strains. The research on the potential developed emulsions based on xanthan-propolis-cinnamon for skin inflammations therapies by applying on textile materials is in progress, studying the potential of biocompatibility by skin irritations potential.

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