ANTIMICROBIAL PROPERTIES OF INSOLES PRINTED WITH MODIFIED BENTONITE AND EXTRACT OF *PICEA OMORIKA*

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ABSTRACT: *In order to remove harmful microorganisms, textile materials are subjected to various types of antimicrobial treatments. Skin infections represent a permanent diagnostic and therapeutic challenge, and can be caused by bacteria, viruses, fungi and parasites. In this paper, the impact of printing insoles with alginate paste, modified bentonites and Picea omorika plant extract on their antimicrobial, dielectric and sorption properties was investigated. It was found that the printed insoles show a certain antimicrobial effect on the bacteria Staphylococcus aureus and Escherichia coli and the yeast Candida albicans. The obtained values of specific conductivity are in agreement with the tested parameter of the sorption properties of the insoles.*

Key words: *insoles, bentonites, Picea omorika, antimicrobial and dielectric properties.*

ANTIMIKROBNA SVOJSTVA ULOŽNIH TABANICA ŠTAMPANIH MODIFIKOVANIM BENTONITOM I EKSTRAKTOM *PICEA OMORIKA*

APSTRAKT: *U svrhu uklanjanja štetnih mikroorganizama tekstilni materijali podliježu raznim vrstama antimikrobnih obrada. Infekcije kože predstavljaju trajan dijagnostički i terapijski izazov, a mogu biti uzrokovane bakterijama, virusima, gljivicama i parazitima. U ovom radu istraživan je uticaj štampe uložnih tabanica alginatnom pastom, modifikovanim bentonitima i ekstraktom biljke Picea omorika na njihova antimikrobna, dielektrična i sorpciona svojstva. Ustanovljeno je da štampane uložne tabanice pokazuju određeno antimikrobno dejstvo na bakterije Staphylococcus aureus i Escherichia coli i* *kvasnicu Candida albicans. Dobijene vrijednosti specifične provodljivosti su u saglasnosti sa ispitivanim parametrom sorpcijskih svojstava uložaka.*

Ključne riječi: *uložne tabanice, bentoniti, Picea omorika, antimikrobna i dielektrična svojstva.*

1. INTRODUCTION

The contamination of textile materials by microorganisms, such as pathogenic bacteria, bacteria that create unpleasant odors, fungi and viruses, is a major concern for public health, as they can cause, at the very least, discomfort, but also skin irritation, hypersensitivity, numerous diseases, and infections in the hospital environment as well as in everyday life [1-3]. Insoles are in constant contact with the foot, so the materials from which they are made should not contain toxic pigments, heavy metals, polyvinyl chloride, chlorofluorocarbon, formaldehyde and harmful volatile solvents, which imposes the need for continuous monitoring of their quality and composition. The most common causes of fungal infections are dermatophytes, aerobic fungi that can enter and infect the keratinized layers of the skin, hair and nails. Skin infections can be caused by bacteria, viruses, fungi and parasites. Some bacteria can cause disease in individuals who have an open wound through which bacteria can enter the body or in individuals with weakened immune functions. Antibiotic resistance generally makes it harder to fight bacterial infections. Even the World Health Organization, in an alarming report on antimicrobial resistance, warned of the danger of entering the "post-antibiotic era", in which frequent infections and minor injuries can be fatal [4].

Antimicrobial agents for textiles include inorganic salts, organo-metallic compounds, iodoforms, phenols and thiophenols, antibiotics, heterocyclic compounds with anionic groups, oxygen compounds, urea, formaldehyde derivatives and amines. Most of these agents have an unfavorable effect on the biosphere, which is why biodegradable and more environmentally friendly antimicrobial agents, including plant extracts, have come into use. In order to improve the effect, compounds or nano-particles of different metals that exhibit antimicrobial properties are added to the extracts, and the most commonly used are silver, copper and zinc. The effect of silver ions on the bacteria *Staphylococcus aureus* and *Escherichia coli* was demonstrated through the separation of the cytoplasmic membrane from the bacterial cell wall [5]. Silver nanoparticles can completely inhibit the growth of bacterial cells, destroy the permeability of bacterial membranes and reduce the activity of some membrane enzymes, which ultimately causes the death of bacteria [6]. The results of experiments performed with ZnO nanoparticles, which were synthesized in diethylene glycol (DEG), confirmed that *E. coli* cells were damaged after contact with DEG and ZnO, due to the disruption of the cell membrane structure of this bacterium [7]. Optimized sterilization efficiency against *E. coli* and *S. aureus* bacteria of 99.4% and 98.0%, respectively, was obtained in research with ZnO/C-$ZnFe_2O_4$ coral structure, which was illuminated with visible light [8]. The effectiveness of textiles with an active antimicrobial effect is based on the principle of diffusion, that is, bioactive substances are dispersed at a

variable rate from the surface of the fibers by the effect of ion exchange, replacing cations from sweat [9].
Reducing the content of unpleasant odors can be achieved with antimicrobial treatments and by reducing the activity of sweat glands. The number of types of bifunctional textiles with antimicrobial activity has increased considerably in the last twenty years [10]. The goals of using antimicrobial finishes are to significantly limit the frequency of bacterial growth, reduce the formation of odors as a result of the microbiological degradation of sweat, and to avoid the transfer and spread of pathogenic microorganisms [11]. Due to the fact that biologically active substances from plants can slow down or prevent the growth of microorganisms, there is a growing interest in studying their application in textile processing [12,13]. Colored solutions are usually used for dyeing textiles, which give the textile product a certain degree of coloration, as well as antimicrobial and deodorizing properties, UV protection, etc. [14,15].
For the antimicrobial treatment of textiles with extracts of medicinal plants, plants with deodorizing properties and a beneficial effect on the skin were selected for research, where special emphasis was placed on plants with a high content of bioactive components that alleviate sweat allergies. In this paper, the medicinal plant *Picea omorika* was selected, whose extract shows antimicrobial and deodorizing properties.
Picea omorika is an evergreen tree with a pleasant smell. The extract is obtained from spruce needles, which are 1 to 2 cm long and up to 2 mm wide, flattened with two characteristic bluish-white stoma stripes [16]. Female spruce cones are purple-brown in color and are located at the top of the tree. In the mature state, they reach a length of up to 6.5 cm [17]. Medicinal and antimicrobial components are extracted from these plants, as well as essential oils, which are further used for various purposes. The content of phenols and flavonoids affects the effectiveness of plant extracts in removing fungi, bacteria and viruses.
Bentonite is a type of highly plastic clay that has the ability to expand and turn into a gel when it comes into contact with water. The two basic types of bentonite are sodium bentonite and calcium bentonite. The main difference between these two types of bentonite is that sodium bentonite has the ability to expand while calcium bentonite does not. By modifying bentonite, the specific surface area and total pore volume are reduced, as well as the mean mesopore diameter is shifted towards larger diameters. Zn-modified bentonite showed a better antibacterial effect than Cu- and Na-bentonite samples, while Cu/2Zn had the highest antibacterial effect among Cu/Zn bentonite samples. Cu/2Zn was integrated on non-woven textiles and knitwear using the screen printing method and showed good antibacterial activity [18]. Zn-modified bentonites have a better antimicrobial effect on Gram-positive than on Gram-negative bacteria [19-21].
Given that the World Health Organization pays more and more attention to the prevention of health protection and that there is a very small number of published works related to the prevention of foot health protection with natural materials, such as bentonites and plant extracts, this work is focused on the antimicrobial treatment of insoles. Therefore, the aim of the work was to find an adequate way of processing insoles for footwear using the screen printing process modified with bentonite and *Picea omirika* extract, in order to obtain a product with certain antimicrobial, dielectric and deodorizing properties.

2. MATERIALS AND METHODS

Picea omorika extract was chosen for the antimicrobial treatment of insoles. The insole SS-WL5H3 (Ovnak, Vitez) with a thickness of 2.8 mm and a surface mass of 76.8 gm-2 is made of polyurethane foam (lower part of the insole) and polyester non-woven textile (upper part of the insole). The appearance of insoles and the research plan is shown in Figure 1.

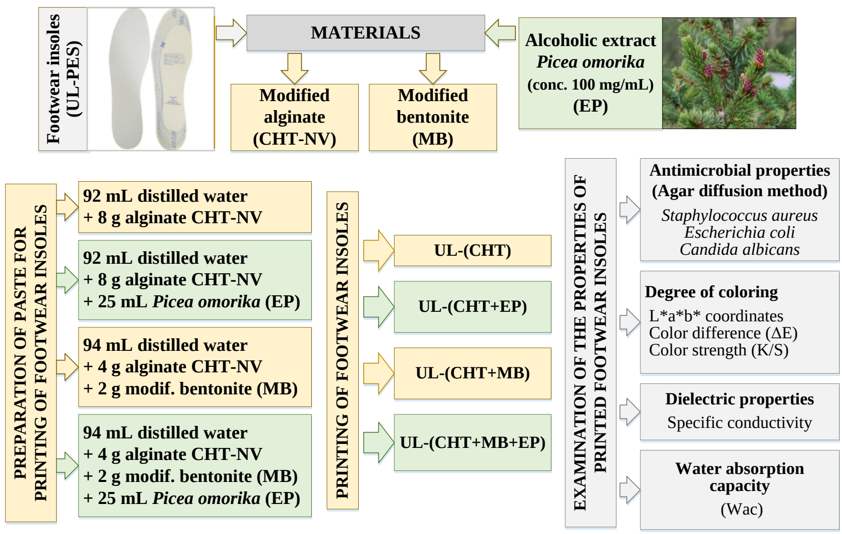


Figure 1: Experiment plan

2.1. Testing of water absorption capacity

The ability to absorb water is expressed by the water absorption capacity (Wac) that the textile material can absorb under defined conditions. The tests were performed according to the DIN 53923 standard [22].

2.2. CIE L* a* b* color coordinates and color intensity measurements

Calculation of the color difference is based on the determination of the differences in coordinates in the color space (ΔL*, Δa*, Δb*) [23,24]. CIE L* a* b* coordinates were determined using a Konica Minolta diffuse spectrophotometer, model CM-2600d. Differences in color (ΔE) between the original insoles and printed insoles were calculated according to equation (1) [25]:

$$\Delta E = \sqrt{\Delta L^2 + \Delta a^2 + \Delta b^2} \quad (1)$$

where ΔL is the color brightness difference between the two samples, Δa is the red/green difference between the two samples, and Δb is the yellow/blue difference between the two samples. The color strength values (K/S) of the printed samples were calculated using the Kubelk-Munk equation:

$$K/S = (1 - R)^2/2R \quad (2)$$

where R is the obtained reflectance at the wavelength of minimum reflection, K is the absorption coefficient, and S is the light scattering coefficient [26].

2.3. Examination of the antimicrobial effect by the diffusion method in agar

The antimicrobial effect of printed knitwear samples was tested using the agar diffusion method [27,28]. Cultures of the bacteria *Staphylococcus aureus* and *Escherichia coli*, as well as the yeast *Candida albicans*, were used for testing. For this test, the processed textile material was cut into samples with dimensions of 2.5 cm x 2.5 cm. After incubation, the zone of inhibition was measured.

2.4. Examination of dielectric properties

For all measurements of dielectric properties, an LCR Hameg 8118 device was used, in the frequency range from 20 Hz to 200 kHz, at an average temperature of 21°C. Samples with dimensions of 2 cm x 2 cm were placed between two electrodes. The voltage between the electrodes is 1V. [29]. Two conductance G and susceptance B were measured. The specific values of conductance and susceptance are calculated according to the following formula:

$$G_{spec}(ili\ B_{spec}) = G(ili\ B)x\frac{d}{S} \quad (3)$$

Dielectric permittivity (ε_r) is calculated according to the following formula:

$$\varepsilon_r = \frac{C \cdot d}{\varepsilon \cdot S} \quad (4)$$

where: $C=B/2\pi f$ and $B=B_m-(B_b-2\pi f\varepsilon_0 S/d)$, f is the frequency, C is the capacity, d is the distance between the electrodes, S is the surface of the electrode, ε_0 is the dielectric permittivity of the vacuum.

3. RESULTS AND DISCUSSION

This section presents the results of testing the properties of footwear insoles printed with alginate paste, modified bentonite and *Picea omorika* plant extract. The results of tests on water absorption capacity, degree of coloring, dielectric and antimicrobial properties are presented.

3.1. Results of the water absorption test

The results of testing the water absorption capacity of footwear insoles printed with alginate paste, modified bentonite and *Picea omorika* extract are shown in Figure 2.

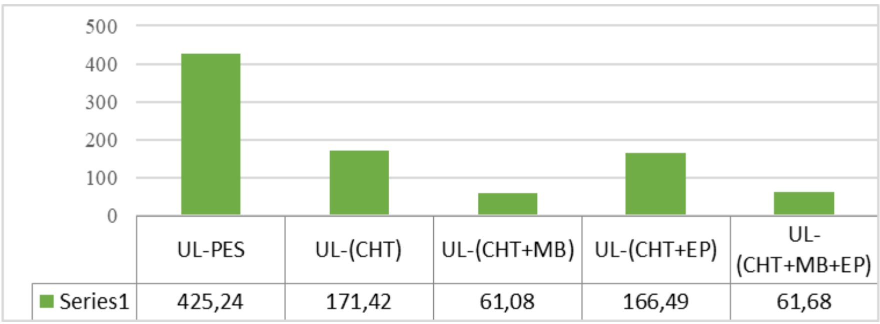


Figure 2: The amount of water absorbed by printed footwear insoles

From the results of the water absorption test, Figure 3.1, it can be seen that the highest Wac was measured in the untreated UL-PES sample (425.24 %), while the lowest Wac was measured in the insole sample printed with alginate paste and modified bentonite UL-(CHT+MB), and was 61.08 %. In general, it can be said that the addition of modified bentonite to alginate printing paste reduces the *Wac* of printed insoles. Also, it can be seen that the addition of *Picea omorika* extract to the alginate paste with modified bentonite had no significant effect on the increase in the amount of absorbed water (*Wac* increased by less than 1%), compared to the UL-(CHT+MB) sample, Figure 2.

3.2. Results of determination of L*a*b color coordinates and color strength

L*a*b* color coordinates of printed footwear insoles, calculated values for color difference (ΔE) and color strength (K/S) are shown in Table 1.

Table 1: L*a*b* color coordinates, calculated color difference (ΔE) and color strength (K/S)

Sample labels	L*	a*	b*	ΔE	K/S
UL-PES	86.79	-1.37	0.50	/	1.72
UL-(CHT)	84.35	-1.22	4.15	4.39	1.80
UL-(CHT+MB)	83.31	-4.44	4.38	6.05	1.75
UL-(CHT+EP)	76.69	-1.51	14.40	17.18	4.02
UL-(CHT+MB+EP)	75.01	-2.66	15.37	19.01	3.44

On the basis of reflectance measurements at wavelength range from 360 nm to 740 nm, spectral curves of printed insoles were obtained, Figure 3, which confirm the results for K/S.

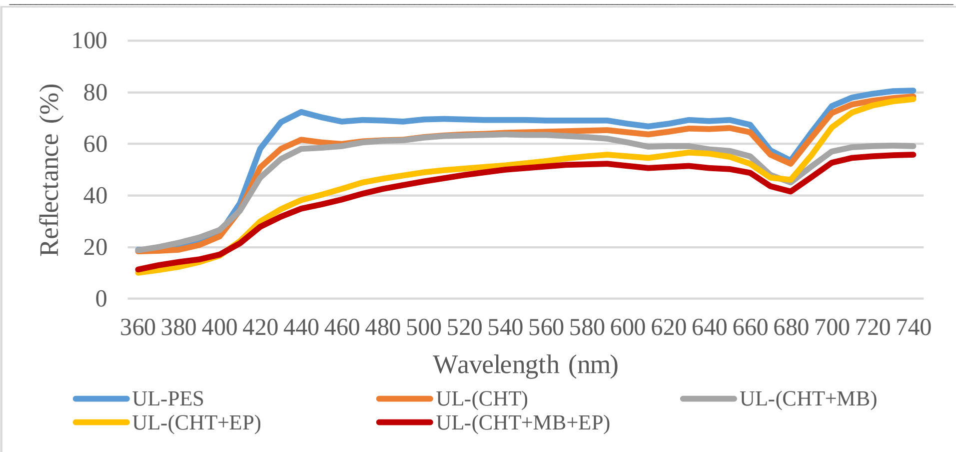


Figure 3: Spectral curves of printed insoles

Based on the measured values of L*a*b* coordinates and calculated color differences (ΔE), Table 1, it can be seen that the samples of footwere insoles printed with alginate paste, modified bentonite and *Picea omorika* extract UL-(CHT+MB+EP) have the biggest difference in color (ΔE = 19.01), compared to the initial sample (UL-PES).
The calculated values (K/S), Table 1, show that the color strength is the highest in the sample printed with alginate paste and *Picea omorika* extract UL-(CHT+EP). By adding modified bentonite to the alginate paste, the K/S value decreases, so K/S for sample UL-(CHT+MB+EP) has a 14.4% lower value compared to sample UL-(CHT+EP).
Spectral curves for insoles, Figure 3, show that the initial sample (UL-PES) has the highest value of reflectance, which is in agreement with the measured color coordinate L* (86.79). This means that the untreated UL-PES sample is the lightest and has the lowest K/S value (1.72), while the most intensely colored UL-(CHT+EP) samples have the highest K/S (4.02).

3.3. Results of dielectric properties

Using the method of dielectric spectroscopy, the conductance and susceptance were measured for all samples in the frequency range from 20 Hz to 7.5 kHz, at a measuring voltage of 1 V. The measurements were obtained at a temperature of 21°C, and the results were processed in the Origin program and graphically shown on the Figures 4 and 5.

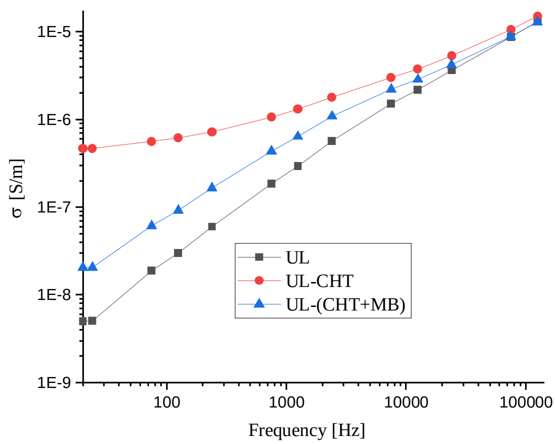


Figure 4: Specific conductivity as a function of frequency for samples UL, UL-(CHT) and UL-(CHT+MB)

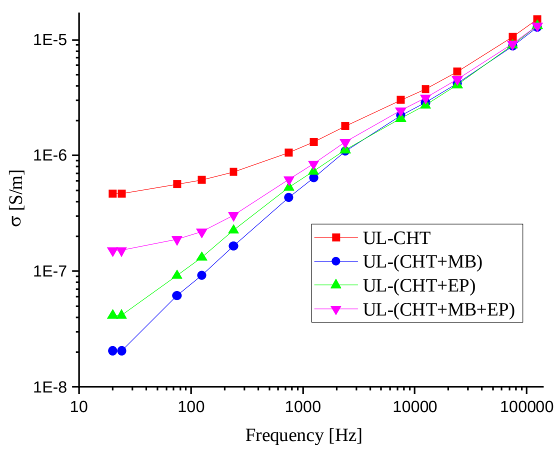


Figure 5: Specific conductivity as a function of frequency for samples UL-CHT, UL-(CHT+EP), UL-(CHT+MB) and UL-(CHT+MB+EP)

Based on the results of specific conductivity (σ) as a function of frequency for samples UL, UL-(CHT) and UL-(CHT+MB), Figure 4, it can be seen that the lowest specific conductivity has the initial sample UL-PES. Also, it can be observed that there was a decrease in specific conductivity when modified bentonite was added to the alginate paste, i.e. the values of σ are lower for the UL-(CHT+MB) sample compared to the UL-(CHT) sample. The obtained values of σ are in agreement with the water absorption capacity (*Wac*).

Sample printed with alginate paste and modified bentonite UL-(CHT+MB) has the lowest specific conductivity. By adding *Picea omorika* extract in alginate paste with modified bentonite UL-(CHT+MB+EP), an increase in specific conductivity was observed, Figure 5. In paper [30], fabrics modified with copper ferrite nanoparticles and *P. omorika* extract were examined and it was found that polyester fabrics have a lower specific conductivity than cotton ones.

3.4. Antimicrobial properties test results

The results of testing the antimicrobial effect of insoles printed with alginate paste, modified bentonite and *P. omorika* extracts are shown in Table 2 and in Figures 6 -8, where a) shows a petri dish with a sample and b) after removing the sample.

Table 2: Results of testing the antimicrobial effect of printed insoles for shoes

Sample labels	Antimicrobial activity on microorganisms		
	Staphylococcus aureus	*Escherichia coli*	*Candida albicans*
UL	*NA*	*NA*	*NA*
UL – (CHT)	*NA*	*NA*	*CI*
UL – (CHT+MB)	*Zi = 0.25 mm*	*CI*	*CI*
UL – (CHT+EP)	*WCI*	*NA*	*CI*
UL – (CHT+MB+EP)	*CI*	*WCI*	*CI*
WCI – weak contact inhibition (25% to 50% - reduction of microorganisms under the sample); PCI – partial contact inhibition (more than 50% - reduction of microorganisms under the sample); CI – contact inhibition (100% - reduction of microorganisms under the sample); Zi – zone of inhibition (mm); NA – no activity			

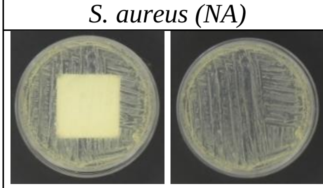
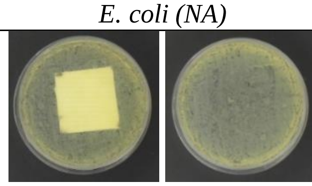
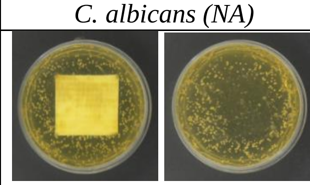
UL		
S. aureus (NA)	*E. coli (NA)*	*C. albicans (NA)*
		
a) b)	a) b)	a) b)

Figure 6: Examination of antimicrobial properties of insoles UL – PES

From the results of testing the antimicrobial properties of insoles, Table 2. and Figure 6, it can be seen that the insole SS-WL5H3, i.e., the starting sample labeled UL-PES, does not show any activity against the bacteria *S. aureus* and *E. coli*, as well as against the yeast *C. albicans*.

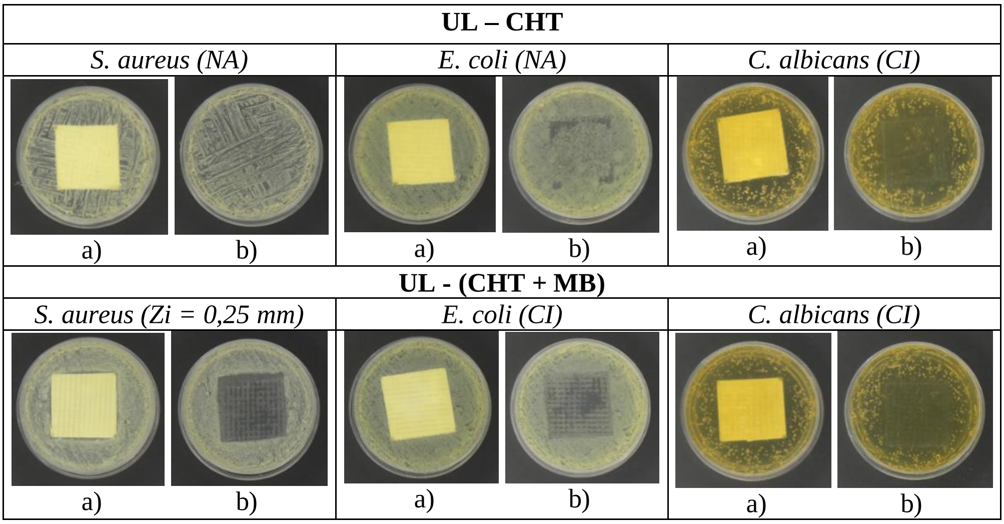


Figure 7: Examination of the antimicrobial properties of insoles printed with alginate paste and modified bentonite

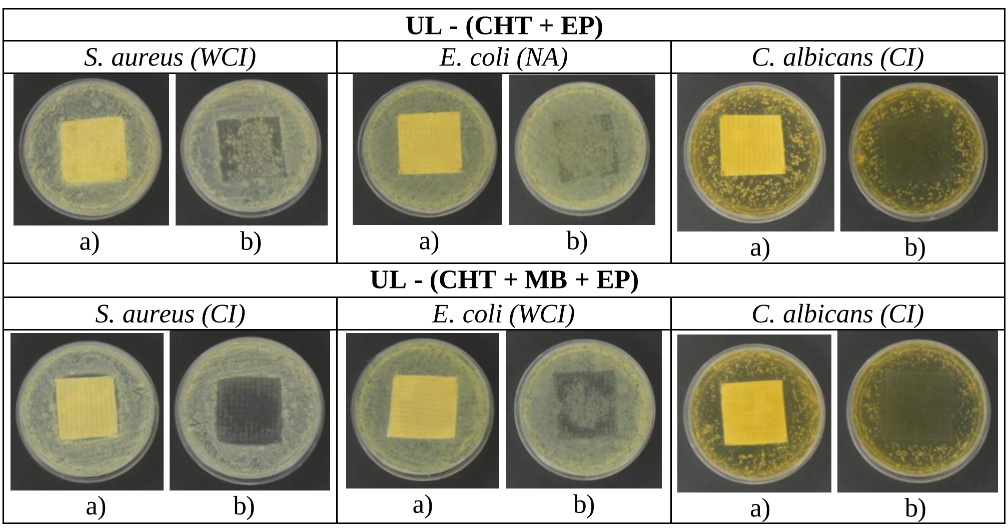


Figure 8: Examination of the antimicrobial properties of insoles printed with alginate paste and modified bentonite with the addition of *Picea omorika* plant extract

Based on the results of testing the antimicrobial properties of insoles printed with alginate paste (CHT-NV) and modified bentonite (MB), Table 2 and Figure 7, it can be concluded that the antimicrobial effect on selected microorganisms (*S. aureus*, *E. coli* and *C. albicans*) improved when modified bentonite was added to the alginate paste.

The antimicrobial effect of printed insoles UL-(CHT+MB) is reflected in the zone of inhibition (Zi = 0.25 mm) on the bacterium *S. aureus* and contact inhibition (CI) on the bacterium *E. coli* and the yeast *C. albicans*. In other studies, the antimicrobial effect of textiles printed with modified bentonite (Cu/2Zn) on bacteria *Escherichia coli*, *Pseudomonas aeruginosa*, *Staphylococcus aureus* and *Bacillus cereus* has been proven, as well as that the antimicrobial effect increases with an increase in the concentration of modified bentonite [18].
For insole samples printed with alginate paste and *Picea omorika* extract UL-(CHT+EP), Table 3.2. and Figure 3.7, a certain antimicrobial activity is observed in the form of weak contact inhibition (WCI) on the bacterium *S. aureus* and contact inhibition (CI) on the yeast *C. albicans*, while it did not show any activity on the bacterium *E. coli*. Also, from the presented results it can be seen that added modified bentonite improved the antimicrobial activity of the sample UL-(CHT+MB+EP), from WCI to CI for *S. aureus* bacteria and for *E. coli* bacteria from NA to WCI, Table 2 and Figure 8.

5. CONCLUSION

By analyzing the results of testing the properties of footwear insoles printed with alginate paste, modified bentonite and *Picea omorika* plant extract, such as the ability to absorb water, color strength, dielectric and antimicrobial properties, the following can be concluded:

- Samples treated with modified bentonite and *Picea omorika* extract significantly reduce the water absorption capacity of insoles (61.68%) compared to the initial sample (425.24%).
- With the addition of modified bentonite to the alginate paste, the K/S value decreases, so the K/S for the sample UL-(CHT+MB+EP) has a 14.4% lower value compared to the sample UL-(CHT+EP).
- At lower frequencies sample printed with alginate paste and modified bentonite UL-(CHT+MB) has the lowest specific conductivity. By adding *Picea omorika* extract in alginate paste with modified bentonite UL-(CHT+MB+EP), an increase in specific conductivity was observed. However, those changes are not observed at higher frequencies.
- Adding modified bentonite increases the reduction of microorganisms under the sample. Samples of insoles printed with modified bentonite and *P. omorika* extract showed a certain antimicrobial effect, mainly in the form of contact inhibition, against all three tested microorganisms (bacteria *S. aureus* and *E. coli* and yeast *C. albicans*).

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