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A DURABILITY ASSESSMENT AND STRUCTURAL CHARACTERIZATION OF BIOPOLYMER-IMPREGNATED WOOD

This paper proposes a mild alternative passive impregnation method of spruce wood with chitosan and zein biopolymers by using a 1-allyl-3-methylimidazolium chloride ionic liquid as a carrier for the impregnant and a swelling agent for the wood structure. The durability of the obtained coatings has been studied by accelerated UV-ageing tests, to provide an insight into the practical applications of the process. The experimental results have indicated relatively high wood percentage gain values for the process, as well as an optimal preservation of the wood surface colour and texture, an increased stability to accelerated ageing determined by the preservation of the values of Brinell hardness, anti-swelling efficiencies and surface energies, in comparison to the non-impregnated reference.

Keywords: passive impregnation, biopolymers, UV-ageing, FTIR spectroscopy, surface energy

Introduction

Increasing the durability of wood coatings is an important requirement for prolonging the life cycle of wood-based products, especially those subjected to demanding environmental conditions, such as exposure to microorganisms, UV, as well as variations in temperature and humidity [Plaschkies et al. 2014; Niklewski et al. 2016]. Due to the strict environmental regulations, nowadays a lot of effort has been made towards developing green coatings based on natural products among which biopolymers are the most promising [Glazkov and Biryukova 2005] However, the limited applicability of this approach, due to the low solubility of biopolymers in traditional solvents, could be mitigated by the use of non-volatile ionic liquids, as we have reported in our previous research [Kosan et al. 2008; Croitoru et al. 2015].

The aim of this paper is to determine the dimensional stability and the durability against UV exposure of Norway Spruce (*Picea Abies*) wood passively impregnated with zein and chitosan biopolymers at 40°C and 60°C, by using

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a 1-allyl-3-methylimidazolium chloride ionic liquid as a mildly ecological carrier for the impregnant. Alkylimidazolium ionic liquids possess low vapour pressures, in contrast with other traditional volatile organic solvents, coupled with good thermal and chemical stability [Garcia et al. 2010]. Ionic liquids have been extensively studied up to date as efficient cellulose and other biopolymers dissolution and functionalization media [Freemantle 2010], as plastifying agents [Ou et al. 2014], antistatic agents [Croitoru et al. 2011a] UV-stabilizers [Croitoru et al. 2011b; Patachia et al. 2012] and antimicrobial agents for wood and cellulose-based materials [Pernak et al. 2004]. While most of the studies reported in the reference literature are focused on the biocidal efficiency of biopolymer coatings [Eikenes et al. 2005], few studies are focused on the dimensional stability of biopolymer-coated wood, on surface characteristics of the coatings following exposure to UV radiation, or on the photodegradation chemistry of chitosan and zein biopolymers.

The performance of the biopolymer coatings submitted for UV-irradiation has been determined by Brinell hardness tests, water mass and dimensional stability coefficients measurements. The structural modifications that occur on the surface of the coatings submitted to accelerated degradative tests have been determined by ATR-FTIR spectroscopy. The modifications in roughness and colouration during the durability tests have been determined by photographic image analysis and the formation of polar groups on the surface of the samples have been determined by surface energy measurements, while the modifications that occur in the microstructure of the impregnated wood and reference have been assessed through optical microscopy tests.

These studies, through detecting the modifications that occur in the chemistry of the impregnated wood exposed to the degradative factors could lead to a better understanding of the mechanisms involved in wood protection, thus leading to the development of more efficient bio-based coatings, with efficiencies comparable to those of synthetic impregnants.

Materials and methods

Materials

The pith-free Norway spruce (*Picea Abies*) wood used for the impregnation and durability testing was cut into rectangular $20 \times 40 \times 0.5 \text{ mm}^3$ specimens and conditioned for 15 days at 25°C and 65% relative humidity, until moisture equilibrium was reached. The average density of the wood was 465 kg/m^3 .

The ionic liquid used in this study, namely 1-allyl-3-methylimidazolium chloride (AMIMCl) with a 99.5% purity was purchased from IoLiTec Ionic Liquids Technologies GmbH, Germany, and used without further purification. The impregnants, namely chitosan (CHS, $M_w = 80\text{-}120 \text{ kDa}$ and 98% purity) and zein (ZE; 95% purity and $M_w \sim 28 \text{ kDa}$) were purchased from Sigma-Aldrich.

Wood impregnation

The impregnation of the wood samples was achieved through (1): immersion of the wood samples for 30 minutes in 20 mL of a 5%_{wt} chitosan and zein biopolymers solution in an AMIMCl ionic liquid at 40°C and 60°C, respectively, followed by (2): the precipitation of the biopolymer into the wood structure through removing the ILs by soaking the wood in distilled water and (3): drying the impregnated materials at 105°C and conditioning them at 65% relative humidity for two weeks.

The 5%_{wt} biopolymers solution in the respective ILs were obtained by gradually adding the required amount of biopolymer into the IL at 90°C under magnetic stirring.

The impregnated samples were conditioned for 15 days at 25°C and 65% relative humidity until the required moisture equilibrium was reached enabling further testing.

Accelerated UV-ageing testing

The conditioned impregnated wood samples were placed in a UV-irradiator (Bio-Link 254, Viber-Lourimat) and exposed for 24 h in total to a UV radiation of 254 nm, with the irradiance value set at 40 mJ/cm². During the irradiation experiments, the relative humidity in the chamber was 45 ± 5% and the temperature 25 ± 5°C. Several sets of identically impregnated wood samples were submitted for UV irradiation, in order to obtain a set of two irradiated samples for each analysis type and test the reproducibility of the phenomenon.

Colour modifications assessment

Colour changes on the wood samples surface were assessed through photographic image analysis, developed in our previous research.

Photographic images of the irradiated samples were taken with the help of a Sony DSC110 digital camera (3072 × 2034-pixel resolution), under identical lighting conditions. The individual images of the samples were loaded in Adobe Photoshop and the L^* , a^* , b^* parameters using the CIELAB (8 bit) channel were determined in twenty points for each specimen, and the average value was used in further interpretations. In the CIELAB colour space, L^* represents the lightness and varies from 100 (white) to 0 (black) while a^* and b^* represent the chromaticity indexes: $+a^*$ red, $-a^*$ green, $+b^*$ yellow and $-b^*$ blue [Patachia et al. 2012].

The colour differences were calculated using Equation 1–3 and the total colour difference ΔE was calculated from Equation 4 for each sample [Forsthuber et al. 2013].

$$\Delta L^* = L_2^* - L_1^* \quad (1)$$

$$\Delta a^* = a_2^* - a_1^* \quad (2)$$

$$\Delta b^* = b_2^* - b_1^* \quad (3)$$

$$\Delta E^* = \sqrt{\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2}} \quad (4)$$

where subscript 1 denotes the values before and subscript 2 denotes the values after UV-exposure. Positive values of Δa^* describe a red shift, negative values of Δa^* a green shift, while positive values of Δb^* represent a yellow shift and negative values of Δb^* a blue shift for the colour of the irradiated samples, in comparison to the initial samples.

To compute the roughness of the wood samples, the images were processed using the MATLAB software version R2009b, with the help of the Image Processing Toolbox, by applying the following work steps: (a): image histogram equalization; (b): extracting the binary profile of the image and (c): computing the relative average roughness (RA) for the selected images (in pixels) [Croitoru et al. 2011a].

Moisture absorption experiments and dimensional change monitoring

The impregnated and conditioned wood samples were immersed in closed glass recipients containing 50 mL of distilled water and their mass and dimensions (length, width, and thickness) were determined at precise time intervals until a constant value was reached (20 days).

The water absorption at equilibrium (WA) for all the impregnated samples, before and after UV-irradiation and the reference was determined with Equation 5 [Pandey and Nagaveni 2008]:

$$WA = \frac{m_a - m_b}{m_b} \cdot 100(\%) \quad (5)$$

where m_b is the initial mass of the oven-dried sample before water soaking, and m_a is the mass of the sample stored in water, at equilibrium.

The volumetric swelling coefficient S_V was calculated by using Equation (6) [Fejfer et al. 2014]:

$$S_V = \frac{V_2 - V_1}{V_1} \cdot 100(\%) \quad (6)$$

where V_2 is the volume of the wood samples after water soaking and V_1 the volume of the wood before soaking.

With the help of WA and S_V , the anti-swelling efficiency (ASE) and the water repellent efficiency (WRE) of the impregnated wood samples could be calculated, by using Equations 7 and 8, respectively [Dukarska et al. 2015]:

$$ASE = \frac{S_{vr} - S_{vt}}{S_{vt}} \cdot 100 (\%) \quad (7)$$

where S_{vr} represents the volumetric swelling of the non-impregnated reference and S_{vt} the volumetric swelling coefficient of the impregnated samples, before and after UV-irradiation.

$$S_v = \frac{W_{ar} - W_{at}}{W_{at}} \cdot 100 (\%) \quad (8)$$

where W_{ar} is the water absorption of the reference and W_{at} is the water absorption coefficient for the impregnated samples, before and after UV-irradiation.

Brinell hardness determinations

The Brinell hardness test method was performed on the surface of the impregnated samples before and after the UV-irradiation tests and reference non-impregnated wood by indenting the test material with a 10 mm diameter hardened steel ball subjected to a load of 500 N, applied for 10 seconds. The diameter of the indentation left in the test material was measured with a micrometre. The Brinell hardness number is calculated by dividing the load applied by the surface area of the indentation, according to the procedure described in the reference literature [Kurt and Ozcifci 2009].

Wetting behaviour

Contact angle measurements on the surface of the reference and the impregnated wood before and after UV irradiation were performed by using distilled water, glycerol and 1-bromonaphtalene as reference liquids, with the help of an OCA System 20 goniometer, provided by Data Physics Co., Ltd. at 25°C. Five different single drops of test liquid with 4 μ L volume were deposited on different regions of the surface of the same specimen.

The surface energy of the wood samples was calculated using the Lifshitz-van der Waals and Lewis Acid-Base (LW/AB) approach, with the help of the instrument's software. According to this approach, the surface energy (γ) is decomposed into a Lifshitz-van der Waals (γ^{LW}) dispersive component as well as into a polar component – γ^p – with Lewis acid (γ^{p+}) and Lewis base (γ^{p-}) contributions respectively, according to Equation 9 [Mohammed-Ziegler et al. 2004]:

$$\gamma = \gamma^{LW} + \gamma^p = \gamma^{LW} + 2\sqrt{\gamma^{p+} \cdot \gamma^{p-}} \quad (9)$$

The initial contact angle θ_0 , which is the contact angle at the beginning of the wetting process (for $t = 0$) for the test liquids was used in the calculation of surface energy by using a semi-quantitative approach, without considering the surface roughness. The dispersive and polar components of the surface tension

of the test liquids were obtained from the reference literature [Rudawska and Zajchowski 2007].

The relative error of the surface energy determinations is 2% for the total dispersive and polar components of the surface energy. This error also includes the influence of water evaporation on the contact angle determination.

ATR- FTIR spectroscopy measurements

To obtain supplementary information about the tested materials, ATR-FTIR spectra of the conditioned impregnated wood samples and reference were obtained with a Bruker-Vertex 70 spectrometer at a 2 cm^{-1} in the $4000\text{-}600\text{ cm}^{-1}$ interval. The spectra were obtained either on the samples cross-sections to confirm the presence of the impregnant in the wood's structure, or on the surface of the samples

Results and discussion

The photographic images of the impregnated wood samples are presented in figure 1. It can be observed, that the impregnation process tends to slightly increase the roughness of the sample's surface, the increase being more pronounced in the case of the chitosan coating.

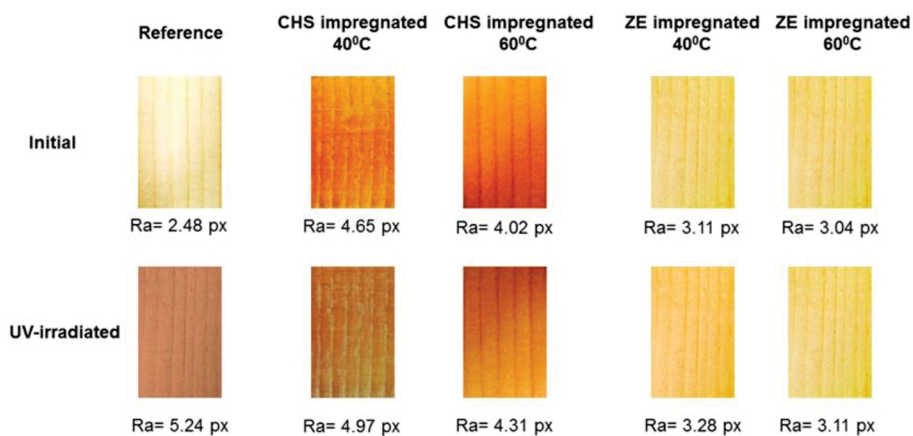


Fig. 1. Photographic image of impregnated samples and reference before and after UV-irradiation

For the zein impregnated samples, due to the lower molecular mass of this polymer and its more pronounced interaction with the ionic liquid carrier, a 64% lower increase in roughness of the samples was attained, in comparison to the CHS-impregnated samples. At 60°C, a lower roughness was attained, than in the case of the 40°C treatment, probably due to the more uniform spreading of

the impregnant on the surface of the wood and its facilitated penetration into the wood mass. The zein impregnated samples preserve the original colour and texture of the wood in a more efficient manner.

After UV-irradiation, an overall modification in the colour of the samples was observed, due to the formation of chromophore groups on the surface of the wood, either by terminal end-groups oxidation or through macromolecular chain scission, in agreement with the FTIR spectroscopy results. The more pronounced colour modifications occur in the non-impregnated reference sample (table 1).

Table 1. Colour parameters of impregnated wood samples and reference before and after UV-irradiation

Impregnated sample		L^*	a^*	b^*	ΔL^*	Δa^*	Δb^*	ΔE^*
Reference	initial	89.5	3.2	6.3	—	—	—	—
	UV-irradiated	45.6	10.4	5.2	-43.9	7.2	-1.1	44.5
Wood-CHS 40°C	initial	65.4	15.8	1.0	—	—	—	—
	UV-irradiated	58.4	25.6	-0.4	-7	9.8	-1.4	12.1
Wood-CHS 60°C	initial	60.5	28.6	-0.8	—	—	—	—
	UV-irradiated	59.6	35.5	-1.2	-0.9	6.9	-0.4	5.1
Wood-ZE 40°C	initial	85.6	4.8	8.6	—	—	—	—
	UV-irradiated	80.2	5.4	9.6	-5.4	0.6	1	5.5
Wood-ZE 60°C	initial	83.5	5.5	8.9	—	—	—	—
	UV-irradiated	82.4	8.9	9.2	-1.1	3.4	0.3	3.5

In the case of the irradiated reference, significant darkening and reddening of the surface were registered, probably due to lignin degradation, as determined from the reference literature. The impregnation of wood with chitosan causes a red shift in the colour of the surface, more pronounced in the case of the samples obtained at 60°C. Zein, in contrast, causes a yellow shift in the colour of the wood surface.

Both impregnants are relatively stable under UV-irradiation, determining a preservation of the surface colour before irradiation. Coating of the wood with zein causes a lower total colour modification parameter, with 8.5-10% than in the case of chitosan at the corresponding impregnation process temperature and with 90% lower than in the case of reference wood.

The WPG of the impregnated wood indicates that the highest percentual weight gain was attained for zein at 60°C, meaning that this compound is able to interact with the ionic liquid carrier and to diffuse through the wood's surface in a more efficient manner than chitosan. Based on the impregnation process parameters, the maximum theoretical WPG that can be attained by using the presented alternative method is in the range of 20-55%, considering that the ionic liquid is used only as a carrier and it is eliminated from the wood in the

precipitation step. For this reason, the WPG values in table 2 are generally lower than those of other traditional processes (50-200%) of impregnation [Hill and Jones 1999; Larnøy et al. 2005; Liibert et al. 2011] where most of the absorbed impregnation solutions or dispersions are fixed into the wood's structure, contributing to the WPG.

Table 2. Performance parameters of the impregnated wood samples and reference

Impregnated sample		WPG (%)	HB (MPa)	WA (%)	S_v (%)	WRE (%)	ASE (%)
Reference	-	—	39.56	110.34	10.11	—	—
Wood-CHS 40°C	initial	23.63	92.67	45.58	5.34	142.61	89.32
	UV-irradiated	—	87.76	56.69	5.97	94.63	69.34
Wood-CHS 60°C	initial	45.67	96.65	28.42	5.36	288.24	88.61
	UV-irradiated	—	95.11	34.54	7.55	219.45	339.05
Wood-ZE 40°C	initial	20.44	56.85	18.74	2.04	488.79	395.58
	UV-irradiated	—	50.45	24.11	2.56	357.65	294.92
Wood-ZE 60°C	initial	56.34	56.98	19.27	1.52	472.59	565.13
	UV-irradiated	—	50.45	20.32	1.86	443.01	443.54

By using the proposed impregnation method, the hardness of the wood could be increased by up to 87% compared to the reference, making the impregnated wood more wear-resistant, resembling other woods with restricted use due to their availability and cost. Chitosan, having the highest molecular weight could be preferentially located at the surface of the wood samples, compared to zein, which diffuses better into the wood's structure. The preferential retaining of chitosan near the surface of the wood generates higher values for the Brinell hardness than the zein-impregnated sample, but, in contrast, zein can retard the water sorption in a more efficient manner, contributing to the stability of the wood samples. The stability to water action is more increased as the impregnation process temperature increases. The accelerated UV-ageing of the impregnated wood samples determines a slight decrease in the values of the anti-swelling efficiencies and water repellent efficiencies, with 2-5% compared to the initial non-irradiated samples, indicating the efficiency of our proposed method.

The impregnation of wood with the dominantly polar chitosan biopolymer seems to slightly increase its wettability, while zein, owing to its hydrophobic character leads to a decrease in the wood wettability (table 3).

The dominating contribution to the polar component of the surface energy of the chitosan impregnated sample is the Lewis base one, since the $-NH_2$ groups of chitosan have electron – donor capacity (Lewis acids) [Johns and Nakason 2011], while zein impregnated wood presents electron-accepting property, which

means that it has a dominating acidic character, probably due to the residual carboxyl groups of serine and isoleucine from zein [Argos et al. 1982].

Table 3. Surface energies of the ionic liquid treated samples

Impregnated sample		$\theta_{0 \text{ water}}$ (degree)	γ (mN/m)	γ^{LW} (mN/m)	γ^p (mN/m)	γ^{p+} (mN/m)	γ^{p-} (mN/m)
Reference	—	33.18	41.22	33.61	7.61	2.22	6.28
Wood-CHS	initial	58.12	37.58	20.3	17.28	2.24	11.14
	UV-irradiated	62.32	41.57	14.0	27.57	1.28	17.25
Wood-CHS	initial	44.63	42.48	17.43	25.05	9.29	16.87
	UV-irradiated	45.74	43.75	14.5	29.25	8.45	18.29
Wood-ZE	initial	34.71	42.68	20.78	21.9	13.54	3.48
	UV-irradiated	35.89	43.25	20.85	22.4	15.41	4.32
Wood-ZE	initial	30.19	40.52	21.66	18.86	12.21	4.18
	UV-irradiated	32.97	40.75	21.97	19.78	16.74	4.28

The surface of the irradiated samples submitted to UV irradiation presents a more hydrophilic character, determined by an increase in the polar component of the surface energies. The most efficient preservation in the surface properties of the coating under UV irradiation is registered for the sample impregnated with zein, at 60°C, probably since at higher temperatures, its intermolecular bonding is disrupted, favouring its interaction with cellulose in a more pronounced manner than in the case of the samples obtained at 40°C. The decrease in the Lewis base parameter for chitosan, respectively the increase of the Lewis acid parameter for zein coatings submitted to UV irradiation could possibly be explained by the chain scission of the macromolecular polysaccharide backbone, respectively the increase in the carboxyl groups' formation through oxidation in the presence of atmospheric oxygen. The lower values of surface energy for zein impregnated samples could be linked to a higher resistance of the surface to water action through minimizing its effective contact surface with the coating.

The main bands in the FTIR spectra of neat wood (fig. 2) are assigned to -OH stretching at $\sim 3325 \text{ cm}^{-1}$, asymmetric methoxy C-H stretching at 2921 cm^{-1} , absorbed water at 1640 cm^{-1} , as well as several bands assigned to different groups of lignin, such as those at 1770, 1740, 1590, 1510, 1270, and 1140 cm^{-1} ; bands common to lignin and cellulose such as those at 1460, 1420, 1375, 1330, 1230, 1160, 1110, and 1030 cm^{-1} ; and bands assigned to cellulose such as those at 1315, 1280, 1180, and 1060 cm^{-1} [Popescu et al. 2009; Wang et al. 2009].

The CHS FT-IR spectrum reveals several distinctive absorption bands at 1662 cm^{-1} (amide I), 1605 cm^{-1} ($-\text{NH}_2$ bending), and 1393 cm^{-1} (amide III). The absorption bands at 1164 cm^{-1} (asymmetric stretching of the $-\text{COO}-\text{OC}$ bridge), 1092 , and 1042 cm^{-1} (skeletal vibration involving the $\text{COO}-$ stretching) are a characteristic of its polysaccharide structure. Several intense bands are characteristic of zein, mainly such as the band at 3200 cm^{-1} , 1650 cm^{-1} and 1512 cm^{-1} assigned to the protein specific amide groups ($-\text{CO}-\text{NH}-$) [Momany et al. 2006; Torres-Giner et al. 2009]. The presence of AMIMCl ionic liquid characteristic bands (such as the sharp discrete absorption in the $1550\text{--}1600\text{ cm}^{-1}$ ascribed to in-plane C–C and C–N symmetric and asymmetric stretching of the imidazole ring) has not been observed in the impregnated samples spectra, due to its removal from the wood structure [Keskar et al. 2012].

The FTIR spectra of the cross-sections of the CHS-impregnated wood samples with AMIMCl, present the characteristic amide I and $-\text{NH}_2$ bending absorption bands of chitosan, thus proving the penetration of this impregnant into the wood structure (fig. 3). Furthermore, shifting of the absorption bands characteristic to wood and chitosan to lower wavenumbers was recorded, due to a flexibilization effect of the cellulose matrix during the impregnation process, as the alkyimidazolium ionic liquids are able to convert the dominantly crystalline cellulose I anomer into the dominantly amorphous cellulose II anomer, as determined from the reference literature and our previous studies [Kosan et al. 2008; Croitoru et al. 2011b].

As expected, at 60°C the relative increase in the intensity of the band at 1652 cm^{-1} is 30% higher than at 40°C , signifying that a higher amount of impregnant could diffuse into the wood structure. In a similar manner, a 45% increase in the relative intensity of the band at 1512 cm^{-1} could be observed in the ZE-impregnated samples, using AMIMCl as a carrier. The presence of this band also in the FTIR spectra of the impregnated samples cross-section proves the penetration of this compound also into the structure of the wood (fig. 3).

Since the area under the infrared band at a given wavenumber is directly proportional to the number of chemical/physical bonds on the sampled volume, ATR-FTIR, for the CHS impregnated sample, the ratios between the area of the band at 1605 cm^{-1} (specific to chitosan) and 1213 cm^{-1} (specific to wood) has been used in order to determine a relative thickness index (I, from fig. 2) [Kane et al. 2009], which reflects impregnant concentration on the wood's surface (the higher the relative thickness index, the higher the thickness of the coating). In the case of zein, the ratio between the area of the bands at 1410 cm^{-1} and 1081 cm^{-1} were used (table 4).

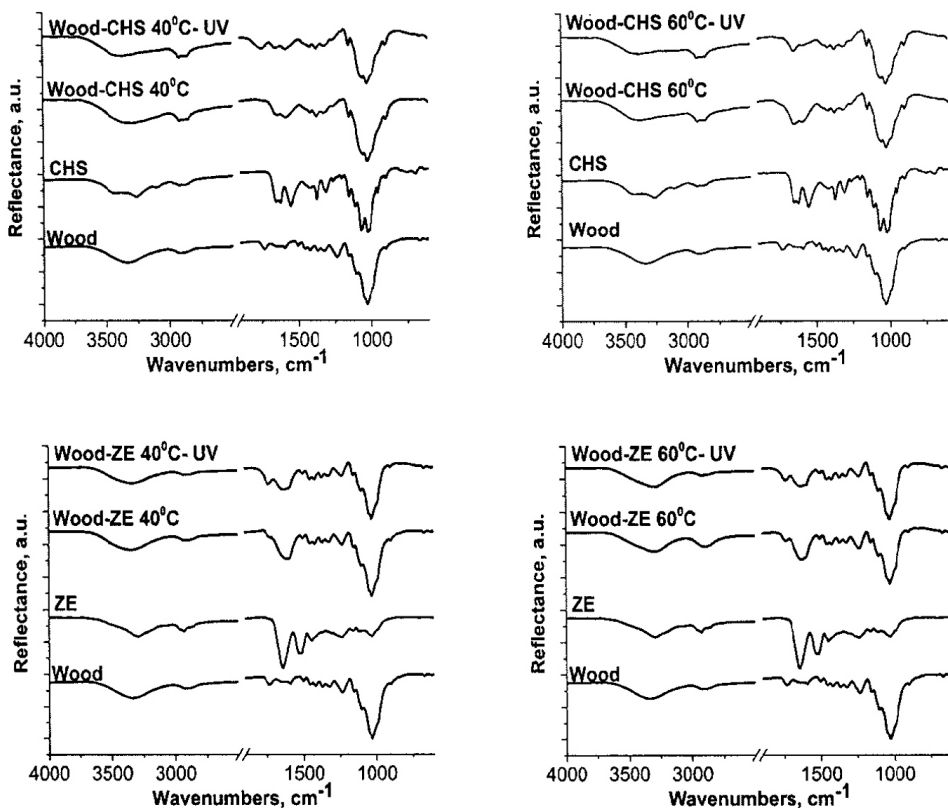


Fig. 2. ATR-FTIR spectra of the initial impregnant-coated surface of the wood before and after UV-irradiation

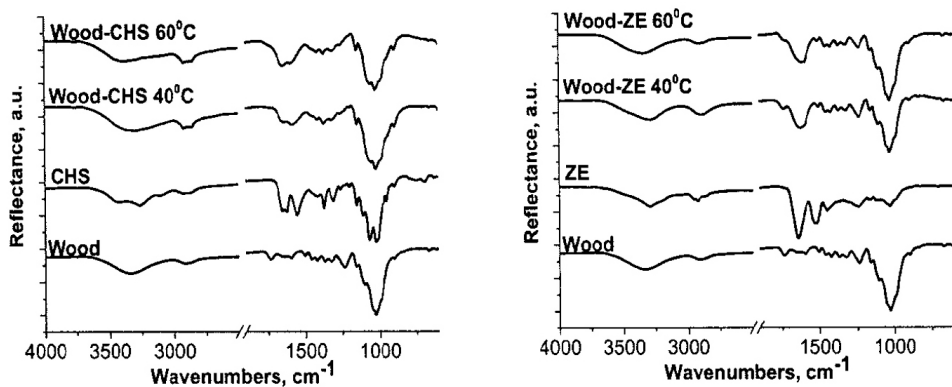


Fig. 3. ATR-FTIR spectra of the chitosan and zein-impregnated samples cross-sections

Table 4. Relative thickness index for the wood coating before and after UV-irradiation

Impregnated sample	I	
	Initial	UV-irradiated
Wood-CHS 40°C	2.36	1.98
Wood-CHS 60°C	1.94	1.92
Wood-ZE 40°C	2.89	2.80
Wood-ZE 60°C	3.41	3.38

It was observed that the higher the temperature of the impregnation process, the higher the index I coating. The structural modifications that occur on the surface of the coatings submitted to the UV-degradative tests fall mainly into three types: (1): flexibilization of the macromolecular chains, depicted through shifting of the characteristic absorption bands of wood and impregnant to lower wavenumbers; (2): an overall decrease of the absorption bands of the impregnant present on the surface of the wood due to the energetically intense UV degradation conditions and (3) an increase in the intensity of the chromophore carbonyl bands, centered at $\sim 1735\text{ cm}^{-1}$.

The chitosan coating obtained with an AMIMCl ionic liquid carrier at 60°C is the most resistant to UV degradation, as determined by the 12-20% lower increase of the relative hydroxyl and carbonyl bands intensity on the surface of the irradiated samples when compared to the non-radiated sample. The most stable coating seems to be obtained with zein impregnant at 60°C, with significant preservation of the surface colour after UV irradiation than in the case of the chitosan coating, as well as lower surface energy, with a dominating dispersive component.

Conclusions

In this paper, the performance of several wood organic biopolymer coatings namely with zein and chitosan, obtained through passive impregnation by using a 1-allyl-3-methylimidazolium chloride ionic liquid as a carrier at different temperatures, namely 40°C and 60°C, was evaluated through accelerated UV-ageing testing.

By our proposed method of impregnation, the natural texture of wood and its natural colour is preserved in a more significant manner than with other traditional impregnation processes. The chitosan and zein coatings are relatively stable under accelerated UV-ageing, as demonstrated by the minimal total colour modification parameters and FTIR spectroscopy. The dominating degradation mechanism of the coatings submitted to UV is the end-groups oxidation and/or macromolecular chain-scission.

The dimensional stability of irradiated and reference impregnated samples were assessed, to emphasise the potential practical applications of the proposed method. The samples of wood impregnated with zein present the highest dimensional stability to water action, having anti-swelling efficiencies with up to 80-150% higher than those registered for chitosan, at the same temperature treatment.

Also, the proposed method of impregnation does not significantly alter the surface properties of wood, a useful feature when designing applications that require the use of water-based primers, adhesives and paints. The surface properties of impregnated wood are well maintained during the accelerated UV-ageing tests.

By the proposed method of impregnation, the hardness of traditional common wood species (such as spruce wood) could be increased to a value comparable to that of exotic wood species, of which usage presents a lower economic potential. The hardness of the coating is maintained during the accelerated UV-ageing tests, only a decrease of 1-3.45% being registered.

The main advantages of the proposed impregnation method are the usage of an ionic liquid carrier which does not pollute the atmosphere, and considerably lower temperatures (40-60°C) than conventional processes of impregnation (100-200°C), which avoids the degradation of the wood material, thus preserving its properties.

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