WOOD RESEARCH doi.org/10.37763/wr.1336-4561/69.2.324336
e-ISSN 2729-8906
69(2): 2024 324-336 pp.

CHANGE IN SOME ACOUSTIC PROPERTIES OF WOODS USED IN MUSICAL INSTRUMENTS AFTER CHEMICAL MODIFICATION WITH PROPIONIC ANHYDRIDE

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(RECEIVED MARCH 2024)

ABSTRACT

The spruce *(Picea orientalis*), maple (*Acer pseudoplatanus*), sapele (*Entandrophragma cylindricum*), cypress (*Cupressus sempervirens*) and mulberry (*Morus alba*) woods, which are among the wood types commonly used in making musical instruments, were subjected to chemical modification by propionic anhydride for 1 h, 3 h and 6 h reaction times. The changes in dimensional stability, sound velocities, modal frequencies and dynamic elasticity modulus values of wood samples after modification were investigated. According to the results obtained, as the weight gain values occurring depending on the reaction time increased, the dimensional stability increased in all wood types. When the acoustic properties were examined, it was determined that individual changes occurred at different values depending on the wood type and chemical modification times.

KEYWORDS: Chemical modification, propionic anhydride, dimensional stability, acoustical properties, FFT analysis.

INTRODUCTION

Wood, as being sourced from a high elastic modulus than relative to low specific gravity, the most important material used in the making of musical instruments as compared to other materials (Ono 1996, Ono and Norimoto 1983, 1984). Also, it is preferred in musical instruments due to aesthetic properties (Fletcher and Rossing 1998). The sound characteristics of the musical instrument vary depending on the type of wood used. Particularly, species of spruce, maple, cyprus, mulberry, cedar, basswood, alder; and the exotic tree species, rosewood and sapele, commonly name (mahogany spp.), are among the most widely used musical instruments. Although the acoustic and mechanical properties of the wood are unique, their major disadvantage being dimensional changes depending on the use in humid ambient

conditions (Bucur 2006). These dimensional changes cause the acoustic properties of wood to be negatively affected. With the increase in moisture, the tuning problem of the musical instrument and the deterioration of the intonation settings constitute a major problem for the musical instruments.

Modification is the process of improving the existing structure of wood and converting its disadvantages into advantages. In the literature, many researchers have tried to improved dimensional stability by subjecting the wood material to various chemical modification processes (Birkinshaw and Hale 2002, Bongers and Beckers 2003, Çetin et al. 2009, Cetin and Ozmen 2001, Hill et al. 2000, Jorissen et al. 2005, Rowel et al. 1993, Rowell 2005, Rowell and Banks 1987). However, studies have shown that the analysis of the changes in the acoustic properties of the wood samples, especially after chemical modification is quite limited (Ahmed and Adamopoulos 2018, Akitsu et al. 1993, Chang et al. 2000, Li et al. 2000, Obataya 2007, Obataya et al. 2001, 2003, Yano and Minato 1993).

Within the scope of this article, chemical modifications of spruce (*Picea orientalis*), maple (*Acer pseudoplatanus*), sapele (*Entandrophragma cylindricum*), cypress (*Cupressus sempervirens*) and mulberry (*Morus alba*) wood samples using with propionic anhydride were studied. Changes in the acoustic and dimensional properties of wood after chemical modification were investigated.

MATERIAL AND METHODS

Material

All wood samples, spruce (*Picea orientalis)*, maple (*Acer pseudoplatanus)*, sapele (*Entandropragma utile)*, cypress (*Cupressus sempervirens)* and mulberry (*Morus alba)* were kindly supplied from Kortürk Kereste Co. Then the test samples, 40 spruce, 41 maple, 41 mahogany, 42 cypress and 39 mulberries, were prepared in the dimension of 220 x 400 x 50 mm to further chemical modifications. Propionic anhydride $(CH_3CH_2CO)_2O$), DMF ($HCON(CH_3)_2$), potassium carbonate (K_2CO_3), toluene, acetone and ethanol were supplied from Merck Chemical Industries (Germany) and used without further purification.

Chemically modification process

Prior to chemical modification, wood samples were subjected to toluene: acetone: ethanol (4:1:1 by volume) extraction for 6 h by using soxhlet extractor, subsequently oven-dried at 103°C over night. The oven-dried weights and sizes of specimens were measured. Several reaction times were determined to modification of wood via propionic anhydride $(CH_3CH_2CO)_2O$ during 1, 3 and 6 h at 110°C and DMF ($HCON(CH_3)_2$) and K_2CO_3 selected as a catalyst. Amount of anhydride to be consumed was calculated according to accessibility of OH^- groups concentration was reported by Hill (2006). It has been found that, 1.134 ml of propionic anhydride and 0.15 g of K_2CO_3 should be used for 1 g of wood material. The propionic anhydride modification was carried out in a lab made stainless steel reactor for 1, 3 and 6 h. At the end of reaction times, chemicals have been depleted from the reactor. And then, subsequent toluene: acetone: ethanol extraction was conducted to remove unreacted anhydride and by-products and oven dried at 103°C over night, then weighted and measured.

The weight percent gains (WPG) of the treated woods were calculated using Eq. 1:

$$\text{WPG}\ (\%) = [(\text{M}_\text{m} - \text{M}_\text{u}) \,/\, \text{M}_\text{u}]\ \text{x}\ 100 \quad (1)$$

where: M_m is the oven-dry mass of the modified wood (g) and M_u is the oven-dry mass of the unmodified wood (g).

Dimensional stability measurements

The modified and unmodified specimens were immersed in distilled water for 5 days at room temperature. The sizes and weights of the specimens were then measured. After measurement, the specimens were oven-dried again at 103°C for 48 h. These measurements were repeated for six cycles to test dimensional stabilities. The swelling S (%) and the anti-swelling efficiencies ASE (%) were calculated by using Eq. 2 and Eq. 3:

$$\text{S}\ (\%) = [(V_\text{ws} - V_\text{od}) \,/\, V_\text{od}]\ \text{x}\ 100 \quad (2)$$
$$\text{ASE}\ (\%) = [(S_\text{m} - S_\text{u}) \,/\, S_\text{u}]\ \text{x}\ 100 \quad (3)$$

where: *Vws* is the water-swollen volume (cm^3), *Vod* is the oven-dry volume (cm^3), S_u is the swelling coefficient of unmodified wood, S_m is the swelling coefficient of modified wood.

Dynamic elasticity modulus

In the determination of dynamic elasticity modulus, free-free flexural vibration method was used (Ono and Norimoto 1983, 1984, Haines et al. 1996). To conduct the experiment, wood samples were placed on two foam prisms at the theoretical nodal point as a supporting (0.224 times the total length of the sample) away from one end of the sample (Fig. 1). A vibration was induced by hitting vertically on the center of sample surface with a metal mallet. A microphone (Apple EarPods standard microphones) was placed at the end of the specimen to acquire the acoustic signal and vibration radiated from the impact. Signal and frequency scans were recorded in MATLAB software. After recording, the acoustic signal was then amplified, filtered and sampled.

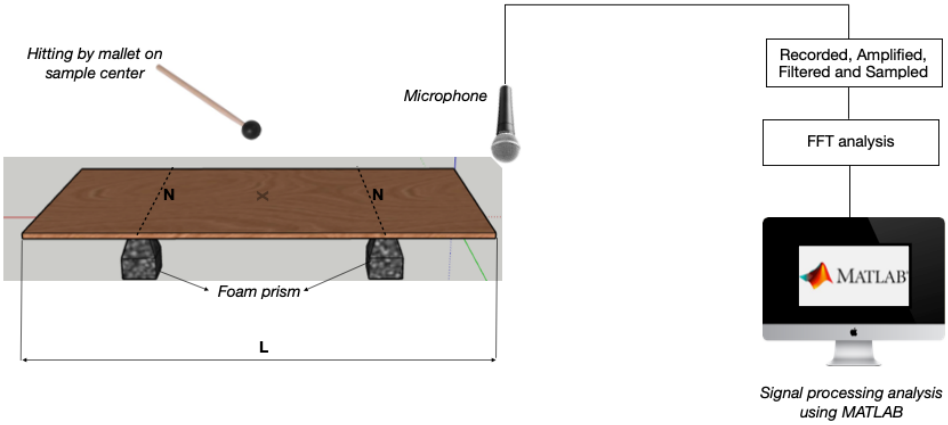

*Fig. 1: Experimental setup for measuring the acoustical properties. N - the nodal point as 0.224 times the total length of the sample (L). N= 0,224 * 400 mm = 89,6 mm.*

The dynamic modulus of elasticity was calculated from first resonance frequency:

$$E_r = \left(\frac{2f_n}{\gamma_n \pi}\right)^2 \frac{m\,L^3}{I} \quad (4)$$

where: m is the specimen weight (g), f_n is the bending vibration frequency (Hz), n is the mode number, L is the length of the sample (mm), γ_n is for the first mode 2.267, and I is the inertia; $I = (bh^3)/12$, where b is the width (mm) and h the thickness of the specimen (mm).

And sound velocity was obtained by Eq. 5:

$$v = \sqrt{\frac{E_r}{\rho}} \quad (5)$$

where: v is velocity (m/s), ρ is the relative density of specimen (kg/m^3).

FTIR analysis

FTIR analysis of chemically modified and unmodified wood were obtained using the KBr (potassium bromide) technique with a Shimatzu 8400s FT-IR spectrometer. The spectra were collected over the 4000 to 400 cm^{-1} wavenumber range, at a resolution of 4 cm^{-1} (40 scans).

Statistical analysis

Analysis of variance (ANOVA) was used to determine the effect of chemically modification on the acoustical and dynamic properties using IBM SPSS software. The resulting F value was compared to the tabular F value at the 95% level of confidence. When F tests resulted in significant differences ($p < 0.05$), to comparisons and identify exactly which groups differ from each other by Tukey as a post-hoc test.

RESULT AND DISCUSSION

Weight per gain (WPG)

The WPG values of different types of wood after chemical modification with propionic anhydride are given in (Tab. 1). As it can be concluded from the results that chemical modification of woods increased the overall WPG values, moreover, it is determined that increased reaction time has further incremental effect on the WPG values. Similar increases in WPG values are observed by Larsson and Simonson (1994) after chemical modification of spruce wood with acetic anhydride as 23.5%. Furthermore, in the study conducted by Li et al. (2000), 25.6% WPG value was obtained after the chemical modification of *Chamaecyparis obtuse* wood samples for 6 h at 110°C with propionic anhydride. On the other hand, Birkinshaw and Hale (2002) reported an increase in WPG values of 16.9, 19.5 and 17.1% for spruce, pine and hybrid woods after modification with acetic anhydride, resp. In addition, 16% WPG values was reported by Popescu et al. (2014) after chemical modification of birch wood with acetic anhydride at 110°C for 6 h. This relatively lower results can be explained by the fact that the acetic anhydride molecules are smaller than propionic anhydride, therefore, lower WPG values could be observed at the similar reaction conditions. On the contrary, similar increases in

WPG values are reported by Çetin et al. (2009) as 26% after a 6 h chemical modification of eucalyptus wood with propionic anhydride. In the study conducted by Rowell and Banks (1987), 22.1% and 14.6% WPG values were obtained in the wood samples for Scots pine and linden woods, respectively, after chemical modification with acetic anhydride for 6 h at 120°C.

Tab. 1: Overview of the acoustical properties and dimensional stability of the samples. Numbers in parentheses represent as a standard deviation.

	N	Velocity (m/s)	Dynamic elasticity modulus (MPa)	Modal frequency (Hz)	WPG (%)	ASE (%)	S (%)
Spruce – unmodified	10	6064.20 *(331,53)*	15471,60 *(933,41)*	627,90 *(11,68)*	-	-	18,79 *(2,43)*
Spruce – 1 h	10	4574,26 *(392,64)*	10697,93 *(1925,63)*	563,30 *(25,80)*	17	42,36 *(2,13)*	10,77 *(1,59)*
Spruce – 3 h	10	4821,97 *(371,95)*	13767,80 *(2302,51)*	612,01 *(20,59)*	24	72,07 *(3,11)*	5,06 *(1,13)*
Spruce – 6 h	10	5544,94 *(388,58)*	17474,37 *(2584,16)*	600,20 *(18,08)*	27	80,98 *(1,53)*	3,29 *(0,19)*
Maple – unmodified	11	4218,09 *(388,05)*	14632,07 *(1544,53)*	509,45 *(16,93)*	-	-	22,20 *(3,13)*
Maple – 1 h	10	5257,45 *(719,89)*	17343,20 *(1223,43)*	459,90 *(52,13)*	13	41,31 *(2,30)*	12,47 *(2,13)*
Maple – 3 h	10	4201,78 *(339,09)*	16347,85 *(1566,05)*	414,10 *(15,96)*	21	62,84 *(4,21)*	8,22 *(1,3)*
Maple – 6 h	10	4000,39 *(185,61)*	13849,58 *(1215,90)*	419,70 *(17,41)*	25	71,95 *(4,21)*	5,57 *(0,84)*
Sapele – unmodified	11	4329,43 *(232,09)*	11004,74 *(1239,49)*	512,27 *(37,13)*	-	-	17,81 *(1,3)*
Sapele – 1 h	10	3973,20 *(204,73)*	10361,66 *(1281,65)*	498,10 *(16,73)*	9	32,63 *(3,21)*	12,35 *(1,98)*
Sapele – 3 h	10	4100,18 *(351,73)*	11208,09 *(1555,05)*	495,20 *(24,15)*	16	42,56 *(3,21)*	11,88 *(1,12)*
Sapele – 6 h	10	4685,89 *(254,03)*	14174,68 *(1527,76)*	506,20 *(12,48)*	21	46,66 *(4,1)*	10,02 *(2,13)*
Cypress – unmodified	12	3826,12 *(666,70)*	13361,4 *(526,90)*	466,60 *(26,46)*	-	-	11,67 *(2,23)*
Cypress – 1 h	10	5025,49 *(231,78)*	15540,32 *(1794,37)*	549 *(18,09)*	9	30,43 *(2,21)*	9,29 *(1,44)*
Cypress – 3 h	10	4788,90 *(261,95)*	15246,61 *(621,96)*	496,2 *(17,37)*	12	57,36 *(4,21)*	5,01 *(0,16)*
Cypress – 6 h	10	4862,91 *(282,22)*	14278,06 *(1867,86)*	519,8 *(15,79)*	23	69,1 *(4,21)*	3,83 *(0,96)*
Mulberry – unmodified	10	3345,50 *(175,96)*	9021,04 *(1021,06)*	443,30 *(9,82)*	-	-	16,48 *(2,43)*
Mulberry – 1 h	10	3520,10 *(262,27)*	10088,60 *(1963,57)*	432,10 *(9,33)*	11	22,0 *(4,21)*	12,42 *(2,91)*
Mulberry – 3 h	10	3453,90 *(245,20)*	9789,30 *(2078,32)*	424,1 *(10,64)*	17	40,9 *(4,21)*	8,76 *(1,13)*
Mulberry – 6 h	9	2940,22 *(465,21)*	10017,87 *(2271,30)*	340,9 *(21,89)*	21	59,0 *(4,21)*	6,79 *(1,01)*

Dimensional changes

After the dimensional stability test of all wood specimens, it was determined that the chemical modification with propionic anhydride decreased the swelling coefficient S (%) due to the increase in weight gain (WPG). According to the obtained data, there is an increase of

anti-swelling efficiency (ASE) due to the increase in the weight gain (WPG) value after chemical modification in all five wood types. After the chemical modification, a minimum of 20% and a maximum of 80% dimensional stability was achieved in the wood samples depending on WPG values (Tab. 1). Similarly, Li et al. (2000) reported 80% ASE values for 30% WPG after chemical modification of hinoko wood with propionic anhydride. In the same manner, Stamm and Tarkow (1946) observed the ASE value between 70-80% for spruce and maple woods after chemical modification with propionic anhydride. Çetin et al. (2009) determined a WPG value of 26% and an ASE value of 83% of eucalyptus wood after 6 h reaction time with propionic anhydride.

Acoustical properties of chemically modified wood by propionic anhydrite

The dynamic elasticity modulus of wood was obtained by placing the resonance frequency calculated by FFT (Fast Fourier Transform) analysis. The results showed that in some cases the acoustical and dynamic elasticity properties (MOE and sound velocity) are increased by propionic anhydrite modification, especially with sapele and cupressus wood (Tab. 1). On the other hand, the velocity and MOE for spruce and morus woods were decreased by modification. Almost no change in sound velocity occurred after modification of maple wood. The results of analyze of variance (ANOVA), statistically significant differences were found in sound velocity of untreated and treated spruces samples (Fig. 3a). It has been determined that sound velocity tend to decrease after the chemical modification process. Similarly, Yano et al. (1993) reported that the sound velocity decreased by 5% in spruce (*Picea sitchensis*) wood after acetic anhydride modification. On the contrary, chemical modification of maple *via* propionic anhydrite has no significant effect on sound velocity (Fig. 3a). It was determined that 1 h modification increased the sound velocity of samples and after 3 h and 6 h sound velocity decreased ($p < 0.05$). Observed results are in line with the Dreher et al. (1964) after 6 h reaction time of maple. It was determined that the 6 h reaction of sapele has significant changes when compared to untreated and 1 h and 3 h treated samples (Fig. 3a). As it can be concluded from Tab. 1, 1 h and 3 h reaction times decreased the sound velocity, however, this detrimental effect is not significant in comparison to untreated samples. Despite the fact that, the increases in 6 h treated samples are statistically significant.

On the other hand, chemical modification of cypress increased the sound velocity of samples when compared to untreated samples (Fig. 3b). It was found that the incremental effect is statistically significant ($p < 0.05$) for all reaction times and the highest increase in sound velocity was observed in 1 h treated samples. However, same incremental effect could not have observed in mulberry samples and chemical modification of mulberry with propionic anhydride did not change the sound velocity for 1 and 3 h reaction time (Fig. 3b). However, prolonged reaction times (6 h) decreased the sound velocity values of mulberry samples and these decreases found statistically significant ($p < 0.05$). Sound velocity varies depending on the moisture and specific gravity of the material (Calegari et al. 2011, Gerhards 1982, Ilic 2001, Moreno et al. 2011, Oliveira and Sales 2006, Sakai et al. 1999, Yang et al. 2015). It is thought that this increase in sound velocity may be due to the decrease in the moisture content of the wood and the increase in its specific gravity after chemical modification. However,

the presence of propyl groups bonded to wood *via* ester bonds changes the sound transmission rate as a negatively in wood (Akitsu et al. 1993, Norimoto and Gril 1993, Yano et al. 1993).

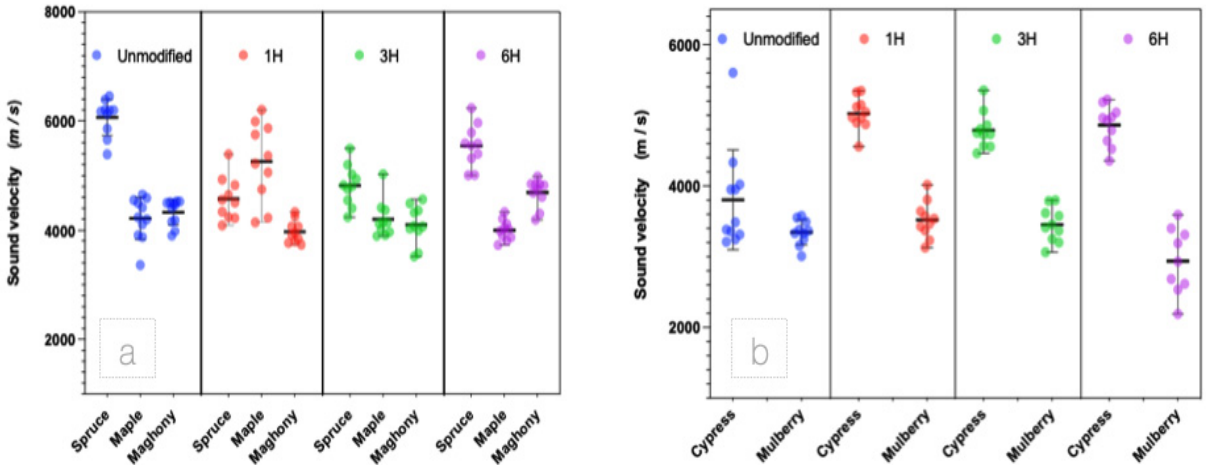

Fig. 4: Sound velocity of the spruce, maple and maghony (a), the cypress and mulberry (b) wood for unmodified and modified with propionic anhydride different reaction time.

Modal frequency

The modal frequency values were compared, there was a numerical decrease in the modal frequency values after the chemical modification process for all wood samples, except cypress wood (Tab. 1). In contrast, after modification, the modal frequency of cupressus wood was increased. For spruce wood, this decrease was found to be statistically significant only in the 1 h and 6 h samples compared to the control samples, while the decrease in the 3 h samples was not statistically significant. In maple samples, the decrease after chemical modification compared to control samples was found to be statistically significant. After the chemical modification, there is a decrease in the modal frequency in maple samples, regardless of the duration of the modification. The decrease in modal frequency was not found to be statistically significant after the chemical modification of sapele wood compared to the control samples ($p > 0.05$). The chemical modification process did not statistically change the modal frequency values of sapele wood. Interestingly, in cypress wood samples, the modal frequency value increased statistically significantly ($p < 0.05$) after the chemical modification process. Among the cypress samples, the highest modal frequency value was detected in the samples that underwent 1 h chemical modification. Mulberry wood was examined, the decrease in modal frequency value did not have a statistical significance in the samples with 1 h modification process. However, this decrease is statistically significant when compared to 3 h and 6 h modified samples ($p < 0.05$). As the modification time increased, the modal frequency of the mulberry wood tended to decrease in a statistically significant manner. The change of modal frequency is rely on the changing of sound transmission rates was due to the formation of carbonyl groups, caused by an extension of the molecular chain in comparison with the original hydroxyl groups (Chang et al. 2000). As a result, the modal properties of sound waves vary. Beside, Yano and Kajita (1994) stated that the frequency values may effected due to the void content in the wood changed after chemical modification. In fact,

Akitsu et al. (1993) stated in their studies that the propyl groups present as bulk in the wood after chemical modification may also have an effect on reducing the resonance frequency.

Dynamic elasticity modulus

A high dynamic elasticity modulus means that the acoustic properties of the material are desirable, especially in musical instrument making. The changes in dynamic elasticity modulus after chemical treatment of wood samples are given in (Tab. 1, Fig. 4). While the dynamic elasticity modulus showed a numerical increase in mulberry wood samples after treatment, this increase was not found to be statistically significant (Fig. 4b). In other words, the dynamic elasticity modulus values did not change statistically after chemical modification ($p > 0,05$). The dynamic elasticity modulus increased significantly all chemically treated samples in cypress wood when compare to untreated one ($p < 0.05$). The highest dynamic elasticity modulus in cypress wood was determined in the 1 h treated samples, also it is found that these changes are statistically significant ($p < 0.05$). However, there is no significance between 3 h and 6 h treated samples ($p > 0.05$). The dynamic elasticity modulus of sapele significantly increased only after 6 h treatment ($p < 0.05$). However, statistically significant difference was not found between control, 1 h and 3 h treated samples ($p > 0.05$). In maple samples, high dynamic elasticity modulus was observed in 1 h chemical treated samples. Furthermore, 1 h and 3 h modified samples had a significant increase when compared to the control samples ($p < 0.05$). After 6 h chemical modification, no significant difference was detected compared to the control samples. In contrary, observed results are decreased 4.3% in Dreher et al. (1964) after 6 h reaction time of sugar maple. In spruce samples, the chemical modification process statistically changed the dynamic elasticity modulus. After the modification, statistically significant decrease observed when compared to the control samples after 1 h and 3 h treatment. Similarly, Larsson and Simonson (1994) stated that small decrease for MOE after acetic anhydride modification of spruce and pine wood. Differently, our 6 h samples were examined, there was an increase, which is not significant, in the dynamic elasticity modulus value compared to the control samples. The highest dynamic elasticity modulus was detected in spruce wood samples after 6h modification.

Yano et al. (1992), Yano and Minato (1992), Minato et al. (2003) found a significant decrease in the elasticity modulus value of sitka spruce wood after the acetylation process. Whilst, Bongers and Beckers (2003) stated that the elasticity modulus values of poplar and Scots pine woods increased after acetylation process, however, similar changes did not observed in beech wood. Besides, Khalil et al. (2010) emphasized that after natural weathering carried out for 1 year, increased to the modulus of elasticity are found in propionic and succinic anhydride modification of mangium wood (*Acacia mangium*) compared to unmodified. On the other hand, Dreher et al. (1964) found that some mechanical properties of Ponderosa pine woods increased after chemical modification of woods with acetic anhydride while, oak and maple wood are decreased. Differently, some researchers stated that the elasticity modulus values did not statistically change after chemical modification (Akitsu et al. 1993, Jorissen et al. 2005, Militz 1991). However, some studies have shown that modification process, especially using anhydride, increases the mechanical properties of the wood as it reduces the moisture content of the wood (Kollman and Côté 1968, Tsoumis 1991). In addition, it is thought that

the increase in density may affect the change in these mechanical properties (Tsoumis 1991). In contrast, some studies have concluded that the chemical modification leads a permanent swelling in the wood. Due to this permanent swelling, the amount of wood polymers decreases in the per unit area, thus mechanical properties are affected in a negative manner (Bongers and Beckers 2003). Besides, chemical modification processes especially carried out in acidic mediums cause by product release in the medium. Since the by products consist of acids (acetic, propionic, maleic), certain amount of degradation occurs in the polymer chains of cellulose, hence resulting in a reduction of mechanical properties. Furthermore, degradation of wood polymers can occur under acidic circumstances (Hon 1996, Hon and Shiraishi 2013, Stamm and Tarkow 1946, Tjeerdsma and Militz 2005).

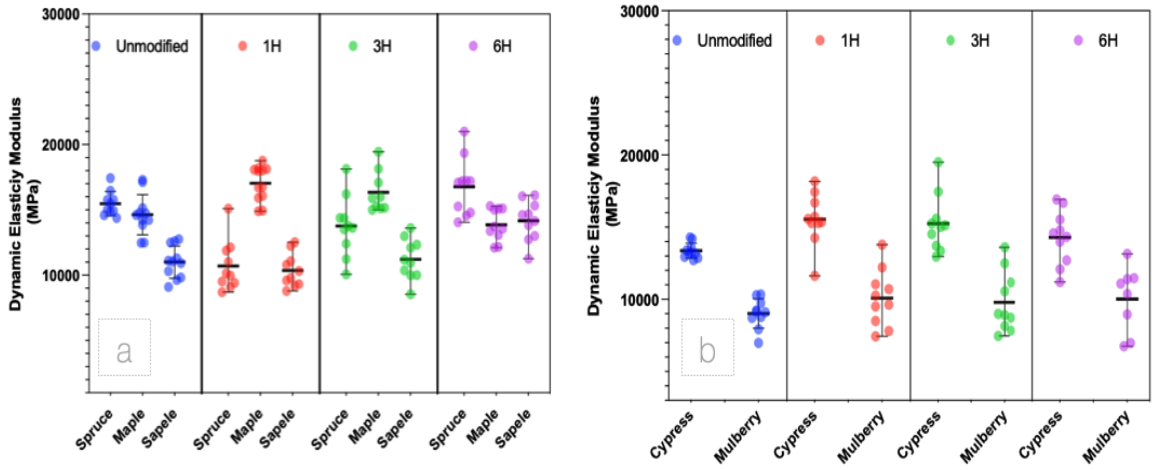

Fig. 4: Dynamic elasticity modulus properties of the spruce, maple and maghony (a), the cypress and mulberry (b) wood for unmodified and modified with propionic anhydride different reaction time.

FTIR analysis

The reaction between propionic anhydride and all wood blocks was characterized by FTIR spectra analysis. FTIR spectra of propionic anhydride-modified and unmodified (control) groups shown in (Fig. 5).

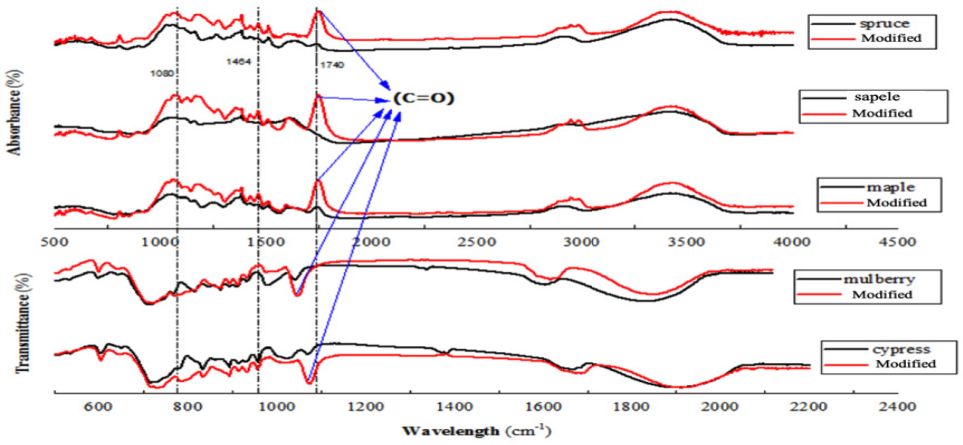

Fig. 5: FTIR spectra of unmodified samples (black) and samples modified by propionic anhydride (red).

The new peak occurring between 1735-1750 cm^{-1} in all five wood types proves that the propyl group is chemically bonded with the unsaturated groups in the wood by forming an ester bond. This new peak is due to the carbonyl groups (C=O) attached to the wood (Silverstein et al. 2005). The peaks occurring between 1300-1000 cm^{-1} originate from carboxyl groups (C-O). The peaks occurring at values of 2987, 1464 and 805 cm^{-1} correspond to the (C-H) bond found in propyl groups and causing sp^3 hybridization (Silverstein et al. 2005).

CONCLUSIONS

The chemical modification process showed different changes in the acoustic and elasticity properties of the wood depending on the type of material and the modification time. Especially for cypress wood samples, the chemical modification process using propionic anhydride had an increasing effect on acoustic and elasticity properties. It was determined that the acoustic properties of sapele wood (commonly name mahogany) increased after the 6 h modification process. However, it has been observed that chemical modification of mulberry wood negatively affects the acoustic properties. Different changes occurred in the acoustic properties of spruce and maple wood depending on the modification time. In terms of elasticity modulus, 1 h and 3 h of chemical modification process with propionic anhydride, caused an increase in the elasticity modulus of maple wood, however, the elasticity modulus of spruce wood decreased within these reaction times. As stated before, the changes that occur after the chemical modification process vary depending on the wood type and reaction time.

Overall, since the dimensional stability of the wood will enhance after chemical modification process, it is thought that it can prevent the tuning problem and the deterioration of the intonation in musical instruments. These topics are planning to fast-growing and non-value-added wood species that have been proceeding to chemical modification may also be the subject of further studies, with the potential to be used in musical instrument making.

ACKNOWLEDGMENTS

This paper has been carried out with financial support from IKCU, BAP-2022-TDR-FEBE-0015.

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