Changes in vibrational properties and colour of spruce wood by hygrothermally accelerated ageing at 95–140 °C and different relative humidity levels

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Abstract

Spruce wood specimens were hygrothermally treated at different temperatures (T , 95–140 °C) and relative humidity during heating (RHh, 0–92%). Their loss in dry mass (ML), specific dynamic Young's modulus (E'/ρ), mechanical loss tangent ($\tan \delta$), and CIELAB colour parameters were measured at 25 °C and 60% relative humidity (RH) before and after the hygrothermal treatment. The changes in physical properties by the hygrothermal treatment were formulated as functions of ML. Those ML dependencies were combined with the ML–time curve at 20 °C previously predicted by using a time– T –RHh superposition, and the changes in physical properties during long-term ageing were predicted. The predicted vibrational properties were stable at 0–80% RH, whereas a significant decrease in E'/ρ and increase in $\tan \delta$ were predicted at 92% RH, suggesting serious degradation in acoustic quality in humid conditions. The predicted changes in colour reasonably agreed with those during natural ageing. The combination of hygrothermal treatment and time– T –RHh superposition is a useful tool to predict the effects of ageing in ambient conditions, particularly when the target property simply depends on the chemical changes in wood constituents.

Keywords Hygrothermal treatment · Ageing · Acoustic quality · Musical instrument

1 Introduction

Hygrothermal treatment is one method used for accelerated wood ageing [16]. Some characteristics of hygrothermally treated wood, such as reduced hygroscopicity, enhanced stiffness, brittleness, and darkened colour [2–4, 7, 13, 14, 17, 18, 20, 26], also appear in naturally aged wood [9–11, 15, 18, 21, 33]. This fact suggests that the effects of long-term ageing can be qualitatively reproduced by hygrothermal treatment.

When hygrothermal treatment is applied as a method to predict the changes in wood properties during ageing, the effects of the relative humidity during heating (RHh) should be considered, because natural ageing occurs at intermediate relative humidity (RH), and the humidity

affects the thermal degradation of wood constituents both quantitatively and qualitatively [8]. However, few investigations have focused on the effects of intermediate RHh [1–3, 6]. In addition, the heating temperature (T) in industrial processing is relatively high (≥ 150 °C), but the chemical reactions induced by heating at ≥ 150 °C are qualitatively different from those at ≤ 130 °C [5]. Thus, the accelerated ageing processes so far proposed, mostly oven-heating, are not sufficient for the precise reproduction of aged wood.

In a previous paper [35], we proposed a time–temperature–superposition (TTHSP) relationship to predict the mass loss (ML) of wood after hygrothermal treatment at arbitrary T and RHh. As the TTHSP allowed the prediction of the ML in ambient conditions, the changes in the

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physical properties of wood during ageing could also be predicted if the physical properties of hygrothermally treated wood were formulated as functions of ML. Many studies have already investigated the ML dependence of physical properties [2, 4, 20, 26, 27], but most of them employed oven-heating at relatively high temperatures. Therefore, these results are unsuitable for the prediction of wood properties after ageing at ambient temperature and intermediate RH.

Based on these considerations, we focus on the vibrational properties and colour of spruce wood because (1) the spruce wood is widely used in the soundboards of musical instruments, (2) vibrational properties and colour are important factors affecting the quality of musical instruments, and (3) many instrument makers are interested in the hygrothermal treatment as a method of accelerated ageing.

In this article, we describe the ML dependencies of vibrational properties and colour parameters of spruce wood hygrothermally treated at 95–140 °C and 0–92% RHh, and then the ML dependencies of the physical properties are combined with the ML–time (t) curves at 20 °C calculated by using TTHSP to predict the changes in the physical properties during ageing. The predicted results are compared with experimental results obtained for naturally aged wood, to verify the validity of hygrothermal treatment as a method for practical accelerated ageing.

2 Materials and methods

2.1 Wood specimens

Sitka spruce wood (*Picea sitchensis*) selected for harp soundboard was used in this study. The lumber was cut into specimens measuring 1.6 mm (tangential) × 15 mm (radial) × 120 mm (longitudinal). The average air-dry density (ρ) of the specimens was 439 kg/m³. To remove the effects of the hygroscopic history during seasoning, the specimens were first moistened at 25 °C and 100% RH for at least 5 days, followed by vacuum drying on P₂O₅ at 20–25 °C for 1 week. The specimens were then conditioned at 25 °C and 60% RH for more than 1 month to determine their vibrational properties and colour parameters.

2.2 Hygrothermal treatment

The specimens were previously conditioned at 20 °C and 0%, 33%, 60%, 84%, or 100% RH for more than 1 month and then hygrothermally treated at 95 °C, 120 °C, or 140 °C and different RHh. For the treatment at 120 °C and 140 °C for 1–7 days, an autoclave equipped with a thermocouple and pressure sensor (PHS-B-500KP, Kyowa Dengyo Co.)

was used. The wood specimens previously conditioned at different RH values were placed in the autoclave with various amounts of deionized water depending on the specified RHh, and then the autoclave was tightly closed and heated to the testing temperatures. The temperature and pressure in the autoclave reached the expected levels within 1 h. The RHh was calculated from the water vapour pressure in the autoclave. The specification and performance of the autoclave are detailed in a previous paper [6]. For heating at 95 °C, specimens were treated in a temperature- and humidity-controlled chamber (PHP-2J, ESPEC Co.) for 1–32 days. For the treatment at 0% RHh, some specimens were heated in an air-circulating oven at 140 °C for 12–47 days to achieve sufficient ML comparable to that obtained by heating in moist conditions. Eight specimens were used for each treatment condition. After the hygrothermal treatments, the specimens were immediately cooled to room temperature (20–25 °C) and vacuum-dried on P₂O₅ to determine their absolute dry mass. The ML is defined as

$$\text{ML (\%)} \equiv 100 \times \frac{M_u - M_t}{M_u} \quad (1)$$

where M_u and M_t are the absolute dry masses of the unmodified and hygrothermally treated wood specimens, respectively.

2.3 Moistening treatment

Hygrothermal treatment involves temporary, reversible changes in physical properties due to the rearrangement of wood polymers via so-called physical ageing [6, 23, 24]. Wentzel et al. recently proposed repeated water soaking to eliminate that reversible effect [32], but the water soaking induces the “irreversible” loss of water-soluble decomposition residues, which significantly affect the vibrational properties of wood [34]. Here we define the reversible effects of hygrothermal treatment as *temporary changes in the physical properties of wood due to hygrothermal treatment, recoverable by moistening at room temperature and 100% RH, involving no irreversible loss of wood constituents*. Based on this definition, the wood specimens were moistened at 25 °C and 100% RH for more than 1 month prior to the measurements of vibrational properties and colour parameters.

2.4 Vibration test

The specific dynamic Young's modulus (E'/ρ) and mechanical loss tangent ($\tan \delta$) of the wood specimens were determined by the free–free flexural vibration method [22], before and after the hygrothermal treatment. The value of E'/ρ was

calculated from the resonance frequency of the first-mode vibration, and that of $\tan \delta$ was determined by approximation of the resonance curve with a theoretical equation. The vibration test was conducted in a chamber where the temperature and humidity were kept at precisely 25 °C and 60% RH, respectively.

2.5 Colour measurement

The CIELAB colour parameters (L^* , a^* , b^*) of the edge-grain surface of the untreated and hygrothermally treated wood specimens were measured with a spectrophotometer (UV-3100PC, Shimadzu Co.) using a D65 light source and an observation angle of 10°. The rectangular sensor head of the spectrophotometer had dimensions of 13 mm × 30 mm. Five specimens were tested for each treatment condition, and measurements were taken at three locations in each specimen. The average values and standard deviations were calculated from the 15 sets of data measured for each treatment condition.

3 Results

3.1 Normalization of experimental values

Table 1 lists the physical properties of 552 tested specimens in the unmodified state. Because wood is a natural material, its physical properties vary to some extent even in the same lumber. In the past studies on the thermal treatment of wood [13, 14, 20], the physical properties of treated specimens were compared with those of the other specimens remaining untreated. In that case, the results are influenced by the original variations in wood properties. To eliminate such variations, we repeated the non-destructive tests on the same wood specimen before and after the hygrothermal treatment, and the physical properties of each treated specimen are normalized according to those in the unmodified state; the obtained relative values are hereafter used to discuss the effects of hygrothermal treatment.

3.2 Effects of heating temperature

We selected the RHh values of 35%, 60%, 85%, and 95%, but the actual RHh values in the autoclave were slightly deviated from those target RHh values, probably because of moisture sorption and desorption of the wood during heating.

Therefore, hereafter we use the experimentally determined average RHh values of 35%, 63%, 81%, and 92%.

In general, the physical properties of hygrothermally treated wood depend on ML, irrespective of T [2, 20, 26]. As examples, the relative values of E'/ρ and L^* at 81% RHh are plotted against ML in Fig. 1. The E'/ρ and L^* values are expressed by single functions of ML, regardless of T . Similar trends are recognized for the other physical properties of EMC and $\tan \delta$, irrespective of RHh. Thus, as far as we discuss the ML dependencies of the tested physical properties, we do not consider the effects of T .

3.3 Changes in EMC

EMC is an important factor to discuss the physical properties of wood, because the physical properties of wood generally depend strongly on EMC. Figure 2 shows the relative EMC of hygrothermally treated wood as a function of ML. The EMC decreases with increasing ML, and the largest decrease is achieved at intermediate RHh. This particular effect of intermediate RHh was previously suggested by Borrega and Kärenlampi [3].

On heating, hygroscopic wood polymers such as hemicelluloses are depolymerized and partly lost. That is a

Fig. 1 Effects of ML on the relative E'/ρ and L^* values of wood hygrothermally treated at different temperatures and 81% RHh. Filled plots, relative E'/ρ ; open plots, relative L^* ; circles, treated at 95 °C; triangles, treated at 120 °C; squares, treated at 140 °C; bars, standard deviations

Table 1 Average values of physical properties of 552 unmodified wood specimens tested

ρ (kg/m ³)	EMC (%)	E' (GPa)	E'/ρ (km ² /s ²)	$\tan \delta \times 10^3$	L^*	a^*	b^*
439 (27)	11.0 (0.1)	14.0 (2.5)	31.8 (4.4)	7.3 (0.5)	80.3 (0.9)	7.3 (0.3)	24.1 (0.7)

Values in parenthesis indicate standard deviations

Fig. 2 Relative EMC values of hygrothermally treated wood as a function of ML. Crosses, treated at 0% RHh; triangles, treated at 35% RHh; diamonds, treated at 63% RHh; squares, treated at 81% RHh; circles, treated at 92% RHh; bars, standard deviations; lines, values approximated by using Eq. 2 for the indicated RHh

major reason for the reduced hygroscopicity and improved dimensional stability of hygrothermally treated wood [8, 28]. By heating in moist conditions, the crystallization of amorphous cellulose [4] can also reduce the EMC because the crystallized cellulose is less hygroscopic than amorphous cellulose. However, some hemicelluloses are seriously depolymerized into low molecular weight sugars at high RHh, and deliquescent compounds remaining in the wood cell wall enhance the EMC of wood [25]. These different changes are combined to give different EMC–ML curves depending on RHh.

3.4 Changes in vibrational properties

Figure 3 shows the changes in E'/ρ due to hygrothermal treatment as a function of ML. The shape of the E'/ρ –ML curve varies widely depending on RHh. At 0% RHh, the E'/ρ is decreased monotonically with increasing ML. At intermediate RHh (35–65%), the E'/ρ increases up to 2–3% ML; thereafter, it decreases monotonically with further increase in ML. Such E'/ρ peaks are also recognized at higher RHh ($\geq 81\%$), but further decreases in E'/ρ are enhanced by elevating RHh.

When the ρ of wood does not vary widely, the E'/ρ value does not depend on ρ but rather on the rigidity of the wood cell wall, which is determined by the volume fraction of rigid crystalline cellulose, the rigidity of amorphous matrix polymers, and the EMC [22]. The E'/ρ is expected to decrease with the depolymerization of hemicelluloses, while it can be increased by the crystallization of cellulose and by decreases in EMC. Since these different effects are

Fig. 3 Relative E'/ρ values of hygrothermally treated wood as a function of ML. See Fig. 2 for definition of symbols

combined, the E'/ρ shows a peak at 2–3% by heating in the presence of moisture (35–92% RHh). Such E'/ρ peaks cannot be explained by the reduction in EMC, because the EMC does not vary in the low-ML range, as exhibited in Fig. 2. No clear peak appears at 0% RHh, probably because a significant cellulose crystallization does not occur in the absence of moisture [4]. At 92% RHh, the E'/ρ is maximized at 1% ML but decreased remarkably by further increases in ML, because of the significant depolymerization of hemicelluloses in humid conditions.

Figure 4 shows the changes in $\tan \delta$ due to hygrothermal treatment as a function of ML. The $\tan \delta$ remains almost unchanged by heating at 0–63% RHh. At higher RHh, the $\tan \delta$ is increased monotonically with increasing ML, and it is drastically increased at 92% RHh.

The $\tan \delta$ value of wood reflects the volume fraction and viscoelastic properties of the amorphous matrix polymers [22]. Therefore, the $\tan \delta$ is expected to decrease with the crystallization of cellulose, loss of matrix polymers, and cross-linking reactions that reduce the mobility of matrix polymers; all of these changes occur upon heating. It was previously demonstrated that the cross-linking reaction occurs in lignin under hygrothermal treatment [29], although the mechanical contribution of cross-linking has not yet been clarified. On the other hand, the $\tan \delta$ is predicted to increase when the mobility of matrix polymers is enhanced by their depolymerization and by the introduction of low molecular weight plasticizers, such as water and sugars. At low or intermediate RHh, these different effects are compensated to stabilize the $\tan \delta$ upon heating. However, the $\tan \delta$ is increased with increasing ML at high RHh, because the effect of depolymerization becomes dominant with elevated RHh. At 92% RHh, the

Fig. 4 Relative $\tan \delta$ values of hygrothermally treated wood as a function of ML. See Fig. 2 for definition of symbols

Fig. 6 Relative a^* values of hygrothermally treated wood as a function of ML. See Fig. 2 for definition of symbols

Fig. 5 Relative L^* values of hygrothermally treated wood as a function of ML. See Fig. 2 for definition of symbols

Fig. 7 Relative b^* values of hygrothermally treated wood as a function of ML. See Fig. 2 for definition of symbols

remarkable increase in $\tan \delta$ is attributed to the plasticizing effect of decomposition residues, i.e. low molecular weight sugars remaining in the wood cell wall [34].

3.5 Changes in colour parameters

Figures 5, 6, and 7 exhibit the ML dependencies of L^* , a^* , and b^* , respectively. All parameters are decreased with increasing ML overall, although a^* and b^* show peaks at low ML. This trend is qualitatively similar to that recognized in oven-heated wood [17]. Higher RHh seems to induce greater decreases in a^* and b^* , but these RHh

dependencies are smaller than those recognized in the vibrational properties.

3.6 Approximation of ML dependencies of physical properties

As described above, the physical properties of the hygrothermally treated wood depend strongly on ML and RHh. Here we approximate these ML dependencies by the following empirical equation:

$$y = 1 + p_1 \left(1 - e^{-\frac{ML}{q_1}} \right) - p_2 \left(1 - e^{-\frac{ML}{q_2}} \right) \quad (2)$$

where y is the relative value of a physical property, and p and q are coefficients reflecting the increases (p_1 and q_1) and decreases (p_2 and q_2) in that physical property with increased ML. These coefficients are determined by the least squares method, while some parameters are considered independent of RHh for better fitting. Table 2 lists the parameters to approximate the ML dependencies of the examined physical properties, and the results of the approximation are exhibited in Figs. 2, 3, 4, 5, 6, and 7 as curves. The approximation is generally successful, although the correlation coefficient is low ($r < 0.9$) at 92% RHh because of data scattering. By using Eq. (2) and the parameters listed in Table 2, the changes in physical properties due to hygrothermal treatment can be estimated at given values of ML and RHh.

Figure 8 shows the RHh dependence of q values for the approximation of E'/ρ -ML curves. Similar RHh

dependencies are recognized in the p and q values for $\tan \delta$ and the colour parameters. Although the physical meaning of these RHh dependencies remains unclear, the diagram allows the prediction of ML dependencies of physical properties at arbitrary RHh.

4 Discussions

4.1 Prediction of wood properties during ageing

As described above, the effects of hygrothermal treatment on the physical properties of wood are formulated as functions of ML at different RHh. This allows prediction of the changes in wood properties during ageing at 20 °C and different RH.

Table 2 Parameters to approximate the ML dependencies of physical properties

	RHh (%)	p_1	q_1	p_2	q_2	r^a
EMC	0	0	–	0.1560	1.383	0.937
	35			0.3499	3.734	0.917
	63			0.3405	3.458	0.984
	81			0.2977	2.514	0.983
	92			0.1916	1.247	0.694
E'/ρ	0	0.2586	9.375	1	25.94	0.978
	35		8.898		50.95	0.978
	63		6.267		42.59	0.964
	81		3.990		28.30	0.985
	92		1.638		9.286	0.882
$\tan \delta$	0	3.098	38.51	1	9.392	N/A ^b
	35		66.17		17.62	N/A ^b
	63		53.36		14.04	N/A ^b
	81		26.45		8.902	0.947
	92		4.219		1.073	0.926
L^*	0	0	–	0.557	3.288	0.990
	35				2.793	0.957
	63				2.726	0.991
	81				2.567	0.986
	92				2.287	0.974
a^*	0	1.295	0.8614	1.707	7.357	0.703
	35		1.031		8.428	0.943
	63		1.402		7.980	0.906
	81		1.333		6.531	0.919
	92		1.209		4.386	0.925
b^*	0	0.2850	0.3296	1	8.673	0.968
	35		0.4162		9.851	0.948
	63		0.6979		7.320	0.981
	81		0.5958		5.965	0.969
	92		1.042		4.763	0.929

^aCorrelation coefficient between the experimental and calculated values

^b $\tan \delta$ value of hygrothermally treated wood shows no ML dependence

Fig. 8 RHH dependence of q values for the approximation of E'/ρ -ML curves. Open symbol, q_1 value; closed symbol, q_2 value

In a previous paper [35], we proposed a time-temperature-humidity superposition (TTHSP) to predict the ML due to hygrothermal treatment at arbitrary T and RHH. In the low-ML range ($< 14\%$), the ML is approximated by a power function of t irrespective of T and RHH, and the master curve at a reference condition (120°C and 63% RHH) is expressed by the following equation:

$$\text{ML}(\%) = 0.6151 \times t_{\text{ref}}(\text{day})^{0.7778}. \quad (3)$$

This master curve can be shifted to a ML- t curve at arbitrary T and RHH by using shift factors (a_T and a_H). The a_T is a shift factor representing the effects of T and is defined by

$$a_T \equiv \frac{t_T}{t_{\text{ref}}}, \quad (4)$$

where t_{ref} is the test time at a reference T (T_{ref}) and t_T is the time required to yield the same response at the test T . Another shift factor a_H , representing the effects of RHH, is defined as

$$a_H \equiv \frac{t_H}{t_{\text{ref}}}, \quad (5)$$

where t_{ref} is the test time at a reference RHH (RHH_{ref}) and t_H is the time required to yield the same response at the test RHH. The ML- t curve at a specific T and RHH is obtained by replacing t_{ref} in Eq. (3) with $t/a_T a_H$.

The temperature dependence of a_T has been experimentally determined as

$$\ln(a_T) = 13402/T(K) - 34.165 \quad (r = 0.996). \quad (6)$$

At 20°C , the a_T is estimated to be 1.04×10^5 . For a_H values, we use the experimental values listed in Table 3, as previously determined [35].

These shift factors allow us to predict ML at 20°C and different RHH. In this case, the RHH is equivalent to the environmental RH, because the wood is not heated at 20°C . Because the ML dependencies of the physical properties have already been formulated, these are combined to predict the changes in physical properties of wood during long-term ageing at 20°C .

Figure 9 shows the predicted EMC values over 2000 years of ageing at 20°C . The EMC decreases with elapsed time, and higher RH causes faster decreases in EMC. Figure 8 also exhibits the EMC values experimentally determined for aged cypress wood used in old temples [33]. As such religious constructions usually utilize quality lumber and have been well maintained by believers, wood samples from such old temples are suitable to discuss the effects of long-term ageing. Although the experimental values are scattered, the predicted EMC at 35 – 63% RH shows reasonable agreement with those of naturally aged wood.

Figures 10 and 11 show the predicted changes in E'/ρ and $\tan \delta$ at 20°C , respectively. The E'/ρ and $\tan \delta$ are expected to remain unchanged at 0% RH. At 35 – 63% RH, the E'/ρ slightly increases with elapsed time, while the $\tan$

Table 3 Shift factor a_H at tested RHH [35]

RHH (%)	0	35	63	81	92
a_H	7.208	2.031	1	0.656	0.262

Fig. 9 Predicted changes in EMC value during ageing at 20°C . Lines, predicted values at the indicated RHH; filled squares, experimental values of naturally aged cypress (*Chamaecyparis obtusa*) wood at 20°C and 60% RH [33]

Fig. 10 Predicted changes in E'/ρ value during ageing at 20 °C. Lines, predicted values at the indicated RHh; open triangles, experimental values of naturally aged spruce (*Picea abies*) wood at 20 °C and 65% RH [15]; open circles, experimental values of naturally aged cypress (*Chamaecyparis obtusa*) wood [12]; open squares, experimental values of naturally aged pine (*Pinus densiflora*) wood at 25 °C and 60% RH [21]; filled squares, experimental values of specific static Young's modulus for naturally aged cypress (*Chamaecyparis obtusa*) wood at 20 °C and 60% RH [33]

Fig. 11 Predicted changes in $\tan \delta$ value during ageing at 20 °C. Lines, predicted values at the indicated RHh; open circles, experimental values of naturally aged cypress (*Chamaecyparis obtusa*) wood [12]; open squares, experimental values of naturally aged pine (*Pinus densiflora*) wood at 20 °C and 60% RH [21]

δ remains unchanged. This suggests that the acoustic quality of wood is expected to remain constant or be slightly increased during ageing at intermediate RH. On the contrary, a significant decrease in E'/ρ and increase in $\tan \delta$ are predicted at 92% RH. This suggests that the acoustic quality is remarkably degraded in humid conditions. Such

adverse changes in humid conditions should be considered when using an aged wood in soundboards. The age of the wood is less important than the humidity to which the lumber has been exposed when selecting quality lumber for the manufacture of musical instruments.

Figures 10 and 11 also exhibit the experimentally determined E'/ρ and $\tan \delta$ values of aged wood [12, 15, 21, 33]. Several points are for specific static Young's moduli (E/ρ) [33]. The plots are scattered widely; no consistent time dependency is recognized. Such variations are attributed to the natural variations in the mechanical properties of wood. The mechanical properties of wood are influenced by anatomical features such as latewood content and the angles of microfibrils in the wood cell wall, as well as the chemical structure of wood polymers. Therefore, the effects of chemical changes during ageing are masked by the anatomical variations, inducing plot scattering.

The mechanical properties of aged wood are worth investigating because aged wood is necessary for the appropriate maintenance and restoration of old wooden constructions and cultural properties [16, 30, 31]. However, the effects of ageing are not precisely evaluated by comparing the physical properties of new and aged wood samples, because of the broad original variations in the mechanical properties of wood. As a prediction of wood ageing, the curves exhibited in Figs. 10 and 11 may be more reliable than the scattered plots of aged wood, because the same specimens were tested before and after the hygrothermal treatment and the effects of humidity were considered in the present study.

Figures 12, 13, and 14 exhibit the predicted changes in L^* , a^* , and b^* with elapsed time at 20 °C, compared with the experimental values for aged cypress wood [18]. In contrast to the scattered plots of E'/ρ and $\tan \delta$, clear trends are recognized in the experimental values, probably because the colour changes reflect the chemical changes in wood polymers with little dependence on anatomical features. The experimental values of b^* are slightly higher than the calculated values, but those of L^* and a^* agree reasonably with the values calculated at 35–63% RH. This fact suggests that the TTHSP is a useful method to predict the colour changes during ageing.

The colour change due to hygrothermal treatment depends on the wood species, as well as the amount and characteristics of the extractives [19]. However, small amount of the extractives do not significantly affect the colour of hygrothermally treated spruce wood [34]. In addition, the changes in colour of spruce wood by thermal treatment are similar to those of cypress wood qualitatively and quantitatively [18, 34] probably because their colours are not significantly influenced by the pale colour of the extractives. Therefore, the results exhibited in Figs. 12, 13, and 14 may be applicable to the other

Fig. 12 Predicted changes in L^* value during ageing at 20 °C. Lines, predicted values at the indicated RHh; filled circles, experimental values of naturally aged cypress (*Chamaecyparis obtusa*) wood in ambient condition [18]

Fig. 14 Predicted changes in b^* value during ageing at 20 °C. Lines, predicted values at the indicated RHh; filled circles, experimental values of naturally aged cypress (*Chamaecyparis obtusa*) wood in ambient condition [18]

Fig. 13 Predicted changes in a^* value during ageing at 20 °C. Lines, predicted values at the indicated RHh; filled circles, experimental values of naturally aged cypress (*Chamaecyparis obtusa*) wood in ambient condition [18]

coniferous wood species whose colour changes upon hygrothermal treatment and natural ageing are similar to those of cypress and spruce wood.

4.2 Artificial reproduction of aged wood by hygrothermal treatment

As described above, TTHSP is useful to predict the effects of natural ageing in ambient conditions. It also enables the precise artificial reproduction of aged wood. Such an artificial ageing process may be attractive for craftsmen

fabricating antique furniture and restorers who use aged wood to repair old wooden cultural properties. Artificial ageing may also appeal to instrument makers, because it can enhance the E'/ρ of wood without changing the $\tan \delta$ value.

Figure 15 shows an example of artificial ageing: plots of ML versus t at 60–100 °C and 63% RHh. When we must maximize the E'/ρ , the ML should be 2–3%. Therefore, a promising heating condition is at 100 °C and 63% RHh for 30 days. On the other hand, when we wish to reproduce the qualities of antique wood, 10–30 days of heating at 100 °C and 63% RHh can achieve 1–2% ML corresponding to 500 years of ageing.

Finally, it should be emphasized that the present results are only valid for clear (no defect) specimens of a particular wood species (spruce) used for the soundboards of musical instruments. If we need to know the behaviours of the other wood species, the same measurement should be conducted on each wood species. Despite such a limitation, however, the combination of TTHSP and ML dependency of wood properties proposed in this article is a useful option to generalize the fragmental results on the hygrothermally treated wood.

5 Conclusions

Spruce wood was hygrothermally treated at 95–140 °C and different RHh, and the vibrational properties and colour parameters of the treated specimens were formulated as functions of ML. In addition, the ML at 20 °C was predicted by using time–temperature–humidity superposition.

Fig. 15 Predicted ML values due to hygrothermally accelerated ageing at the indicated T and 63% RHh

These results were combined to predict the changes in physical properties during ageing at 20 °C. The predicted changes in vibrational properties suggested that the acoustic quality of wood could be slightly improved by ageing at intermediate RH, whereas it is seriously degraded in humid conditions. The predicted EMC and colour parameters showed good agreement with the experimental values of naturally aged wood in the literature.

Compliance with ethical standards

Conflicts of interest The authors declare that they have no conflict of interest.

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