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PAPER

Effects of oriental lacquer (urushi) coating on the vibrational properties of wood used for the soundboards of musical instruments

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Abstract: In order to investigate the possibility of Oriental lacquer (urushi) as a coating for the wooden-soundboard of musical instruments, the effects of urushi coatings on the vibrational properties of wood were compared to those of conventional coatings. By coating, the dynamic Young's modulus of wood decreased slightly in its fiber direction whereas that in the radial direction increased. The most remarkable changes due to coating were recognized in the internal friction of wood (Q^{-1}), especially that in the radial direction. The effect of the urushi coating on the Q^{-1} of wood was relatively small and very close to those of polyurethane coating used for the soundboard of harp. The viscoelastic and mechanical properties of urushi lacquer films were also similar to those of the polyurethane lacquer film. These results suggested the possibility of urushi as a coating for the harp soundboard. The effects of coatings on the vibrational properties of wood were explained by using a model considering three layers, the uncoated wood, coating layer, and a layer consisting of lacquer and wood cell wall.

Keywords: Oriental lacquer, Wooden-soundboard, Dynamic Young's modulus, Internal friction

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1. INTRODUCTION

In recent years, various natural lacquers are reevaluated for their safety and reproducibility. Urushi, the sap of Japanese lacquer tree (*Rhus verniciflua*), is a natural lacquer which has been widely used in east-Asian countries as the coating and adhesive for wooden-products. A lot of ancient wooden-artifacts have been protected by the excellent indoor durability of urushi coating over thousands of years. The urushi is still preferred today as it symbolizes traditional Japanese beauty for its appearance, so that, the artistic works with urushi coating are generally called "japan" [1]. Recent investigations have clarified that the urushi coating has high resistance to water and chemicals [2], and its mechanical properties are comparable to those of synthetic lacquer coatings [3]. However, high cost and intractability of the urushi lacquer are serious problem for its utilization.

In order to utilize the excellent characteristics of urushi, we focused on the possibility of urushi as a coating for the wooden-soundboard of musical instruments. Most of wooden-instruments are made by hand with much time and many processes. Therefore, the prolonged drying duration and special technique required for the urushi coating may cause few trouble, and an additional cost for urushi coating will not induce serious increase of total cost for making instruments. Thus the coating for the wooden-soundboard seems a possible use left for urushi.

When we try to apply the urushi to the soundboard of musical instrument, it should be confirmed that the urushi is similar to conventional lacquers, in respect of their effects on the vibrational properties of wood. Although the viscoelastic and mechanical properties of urushi have been well investigated [4, 5], few studies have dealt with the effects of urushi coating on the vibrational properties of wood. In this paper, the vibrational properties of wood with urushi coating are compared to those with conventional coatings. Furthermore, the effects of coatings are described with the viscoelastic properties of coatings and wood, by using a model considering the porous structure of wood.

2. EXPERIMENTAL

2.1. Wood Specimens

Sitka spruce wood (*Picea Sitchensis* Carr.) for the soundboard of piano was cut into a size of 3 mm (T, tangential direction) × 15 mm (R, radial direction) × 150 mm (L, fiber direction), and a size of 3 mm (T direction) × 15 mm (L direction) × 100 mm (R direction). The former and latter specimens were used for measuring the vibrational properties of wood in the L and R directions, respectively.

2.2. Lacquers

Three kinds of urushi, clear (C), black (B) and virgin (V) lacquers were used. These are called suki-urushi, kuro-urushi and ki-urushi in Japanese, respectively. As conventional lacquers, we employed a two-package type polyurethane lacquer (Clear No. 200 W21F/F, Nihon-Yushi Co.), a clear lacquer consisting of some synthetic resins and nitrocellulose (LC-11, Gengen Chemical Industry Co.) and a natural oil-based lacquer (Osmocolor 3101, Ostermann & Scheiwe GmbH Co.). The polyurethane lacquer (PU) is used for the soundboard of harp at this time. The clear lacquer (CL) and the natural lacquer (NL) are mainly used for common use. These lacquers were applied to both the LR surfaces of the wood specimens. A brush was used for spreading the C, B and NL lacquers, and the V lacquer was applied with a cloth. The PU and CL lacquers were splayed on the wood specimens. These processes were repeated 1 to 3 times for the C, B and PU lacquers, 3 to 9 times for the V lacquer, and 4 to 8 times for CL lacquer, to form their coatings with sufficient thickness. The NL lacquer was applied 2 to 4 times after sealing with a sealer (Osmocolor 1101, Ostermann & Scheiwe GmbH Co.), but its extremely thin coating layer was not detectable because of its penetration into the wood.

Since the polymerization of urushi lacquer requires high humid condition, the urushi coated specimens were dried in a chamber maintained at 20°C and 80% relative humidity (RH) for a day after each applying. The PU, CL and NL lacquers were dried at a room temperature and uncontrolled relative humidity. Finally, all wood specimens were stored at 25°C and 60%RH over a month before measurement of their vibrational properties.

All lacquers except for NL were also applied to a poly-tetrafluoroethylene plate and dried, to obtain the lacquer films with a thickness from 0.2 to 0.4 mm. These films were cut into strips of 70 × 5 mm, and conditioned at 25°C and 60%RH over two months prior to the viscoelastic and tensile tests.

2.3. Measuring Methods

The dynamic Young's modulus (E) and the internal friction (Q^{-1}) of wood specimens in the L and R directions were determined before and after coating by using the free-free flexural vibration method. The measuring apparatus is schematically illustrated in Fig. 1. A thin piece of 3 × 10 mm iron was attached to the end of a specimen, and the specimen hung by silk threads was vibrated by a magnetic driver. The amplitude of vibration was detected using microphone, and the signal passed through a band-pass filter was observed by a FFT analyzer. The specimen and measuring devices were settled in a closed box in which the condition was maintained at 25°C and 60%RH. The E value of the specimen was calculated from the resonance frequency of the first mode vibration, and its Q^{-1} value was calculated from the peak width of the resonance curve. The resonance frequency ranged from 650 to 850 Hz in the L direction, and 450 to 650 Hz in the R direction.

The E and Q^{-1} values of lacquer films were measured by using a viscoelastometer (RHEOVIBRON DDV-25FP, Orientec Co.). First, the E and Q^{-1} values of films were measured in the frequency range of 1 to 100 Hz and at constant temperatures of −10, 0, 10, 20 and 25°C. Next, the temperature dependence of their E and Q^{-1} values were examined at 11 Hz and in the temperature range of −50 to 150°C with heating rate of 3°C/min. The amplitude of vibration was 25 μm for the urushi and PU lacquer films, and 12 μm for the CL lacquer film.

To determine the tensile strength (σ) and strain at break (ε_{max}) of lacquer films, ten strips for each films were subjected to the tensile test, with an effective span of 45 mm and a loading speed of 10 mm/min.

3. RESULTS AND DISCUSSION

3.1. Effects of Coatings on the Vibrational Properties of Wood

The density (ρ_0), dynamic Young's modulus (E_0) and internal friction (Q_0^{-1}) of the uncoated wood are listed in Table 1. The changes of dynamic Young's modulus (E_c/E_0) and those of internal friction (Q_c^{-1}/Q_0^{-1}) due to coating are plotted against the relative thickness of coatings (t_1/t_0) in Fig. 2. Suffixes 0, 1 and c represent the uncoated wood, coating, and coated wood, respectively. The E values in the R direction increased with coating whereas those in the L direction decreased slightly and linearly with an increase of the coating thickness. The Q^{-1} values of wood, especially those in the R direction, were remarkably enhanced with coating. The effects of coatings on the Q^{-1} values in both directions were analogous. These trends qualitatively agreed with the results already reported [6].

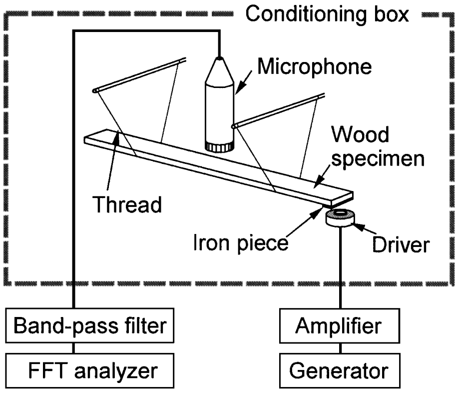

Fig. 1 Apparatus for measuring the vibrational properties of wood specimens.

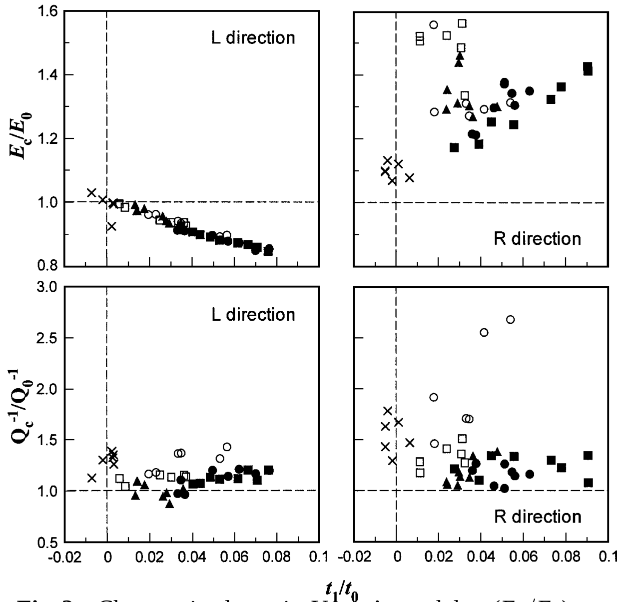

Fig. 2 Changes in dynamic Young's modulus (E_c/E_0) and internal friction (Q_c^{-1}/Q_0^{-1}) of wood specimens in the longitudinal (L) and radial (R) directions due to coating plotted against the relative thickness of coatings (t_1/t_0). ○, Clear lacquer for common use (CL); □, polyurethane lacquer for the soundboard of harp (PU); ×, natural oil-based lacquer for common use (NL); ●, clear oriental lacquer (C); ■, black oriental lacquer (B); ▲, virgin oriental lacquer (V).

Table 1 Characteristics of uncoated wood specimens.

		ρ_0 (g/cm^3)	E_0 (GPa)	Q_0^{-1}
L direction	X	0.442	14.3	0.0068
	SD	0.035	1.18	0.0005
R direction	X	0.436	0.936	0.0206
	SD	0.036	0.230	0.0021

X, average; SD, standard deviation.

The most remarkable effect of the coating was recognized in the Q^{-1} value of wood, especially in its R direction. The Q^{-1} values of wood with CL coating were larger than those with the other coatings, and strongly depended on the coating thickness. This suggested that the Q^{-1} value of CL coating was larger than those of the other coatings. The NL lacquer enhanced the Q^{-1} of wood to some extent while it formed no apparent coating layer. Since it has been confirmed that the NL lacquer constituents could not penetrate into the wood cell wall, the increase in Q^{-1} due to the lacquer should be attributed to its internal coating, *i.e.* the coating on the inner surface of the wood cell walls with the lacquer introduced into the cell cavities. The effects of urushi and PU coatings on the Q^{-1} values of wood were relatively small and independent of their coating thickness. These facts indicated that the Q^{-1} values of urushi and PU coatings were relatively low and comparable to those of wood in the R direction. It should be noted that the urushi coatings are similar to the PU coating, with respect to their effects on the Q^{-1} value of wood which is remarkably affected by coating.

3.2. Viscoelastic and Mechanical Properties of Lacquer Films

As described above, different lacquers give different effects on the vibrational properties of wood, especially its Q^{-1} value. Such variations should be explained by those in the viscoelastic profiles of coatings themselves. In general, the viscoelastic properties of amorphous polymers strongly depend on both the temperature and frequency. Therefore, the effects of temperature and frequency should be taken into consideration to clarify the viscoelastic profiles of the coatings. Figure 3 shows the temperature variations of E and Q^{-1} for the various lacquer films, except for the NL lacquer. Remarkable drops in the E and apparent peaks in the Q^{-1} indicated the glass-rubber transition of the lacquer constituents. Table 2 lists the transition temperature of the lacquer films (T_g) evaluated from the temperature locations of Q^{-1} peaks observed at 11 Hz. Owing to the rigidity of backbone molecules and cross-linking formation between them, the urushi and PU lacquer films recorded higher T_g values than the CL lacquer film.

The variation in T_g affects the viscoelastic profile of lacquer film at a room temperature. Figure 4 shows the frequency variations of E and Q^{-1} for the lacquer films at 25°C. Since the molecular motions in the urushi and PU lacquer films were almost frozen at the room temperature, their E and Q^{-1} varied only slightly with frequency. On the other hand, the E and Q^{-1} of CL lacquer film depended strongly on the frequency.

Table 2 Viscoelastic and mechanical properties of lacquer films.

Lacquer		Density (g/cm^3)	Viscoelastic properties			Mechanical properties at 25°C	
			T_g (°C)[a)]	E (GPa)	Q^{-1}	σ (MPa)	ε_{max}
Synthetic lacquers							
Polyurethane lacquer	(PU)	1.20	110	2.5[b)]	0.050[b)]	38	0.06
Clear lacquer	(CL)	1.18	50	1.4[b)] 1.3[c)]	0.250[b)] 0.260[c)]	1.7	0.02
Oriental lacquers							
Clear (suki-urushi)	(C)	1.15	128	2.6[b)]	0.015[b)]	53	0.06
Black (kuro-urushi)	(B)	1.10	95	2.1[b)]	0.030[b)]	42	0.07
Virgin (ki-urushi)	(V)	1.12	101	2.5[b)]	0.020[b)]	46	0.04

[a)] Temperature location of the Q^{-1} peak detected at 11 Hz.
[b)] Values at 750 Hz evaluated from the frequency dependence at 25°C.
[c)] Values at 550 Hz evaluated from the frequency dependence at 25°C.

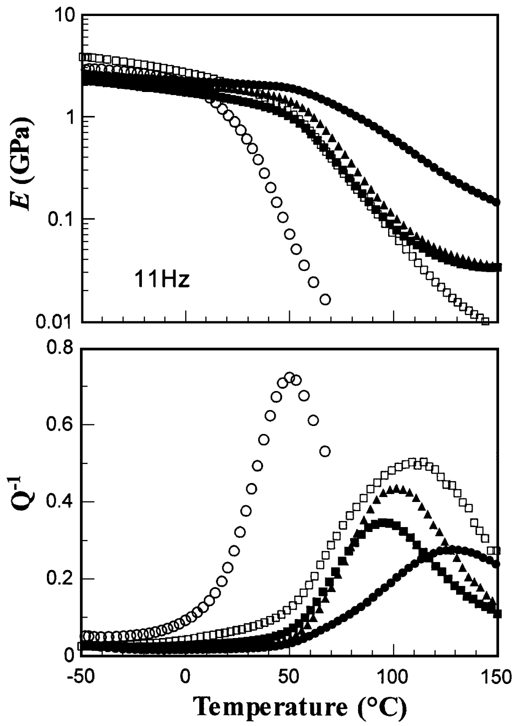

Fig. 3 Temperature variations of dynamic Young's modulus (E) and internal friction (Q^{-1}) at 11 Hz for the lacquer films. Symbols, see Fig. 2.

When the characteristics of lacquer film as a coating for the soundboard are considered, we must clarify its viscoelastic profiles over the audio-frequency range, 20 Hz to 20 kHz, at least. The E and Q^{-1} values of urushi and PU lacquer films at high frequencies could be evaluated from the experimental values determined at low frequencies, because their frequency dependence could be approximated with straight lines in the low frequency range (Fig. 4), and these films showed no apparent relaxation process at low temperatures (Fig. 3). However, such an evaluation was not applicable to the CL lacquer films whose viscoelastic properties depended strongly on the frequency. In this study, the time-temperature superposition theory [7] was adopted to predict the E and Q^{-1} values of CL lacquer films at high frequencies, as its viscoelastic profiles were thermorheologically simple. Figure 5 shows the experimental values of E and Q^{-1} for the CL lacquer films at various constant temperatures (a), and calculated values at 25°C (b). The experimental plots were appropriately shifted along the frequency axis to give a smooth "master curve" at the room temperature.

From these results, we now obtained the E and Q^{-1} values of lacquer films at 550 and/or 750 Hz, as summarized in Table 2. These frequencies are representatives of audio-frequencies and correspond to the average resonance frequency of wood specimens in the L and R directions, respectively. The Q^{-1} values of urushi and PU lacquer films were low and comparable to those of natural wood in the R direction. This fact is consistent with the experimental result that the Q^{-1} values of wood with urushi and PU coatings were relatively low regardless of the coating thickness. On the other hand, higher Q^{-1} values of wood with the CL coating is attributable to high Q^{-1} value of CL lacquer film. Table 2 also lists the mechanical properties of lacquer films determined by the tensile test. The urushi lacquer films recorded high σ and ε_{max} values comparable to those of PU lacquer film, whereas low σ and ε_{max} values of CL lacquer films indicated its fragile nature.

Generally speaking, the viscoelastic and mechanical properties of urushi lacquer films are similar to those of PU lacquer film. It must be remembered that the effects of urushi coating on the Q^{-1} of wood is similar to those of PU coating. These results suggest the possibility of urushi as a coating for the soundboard of harp.

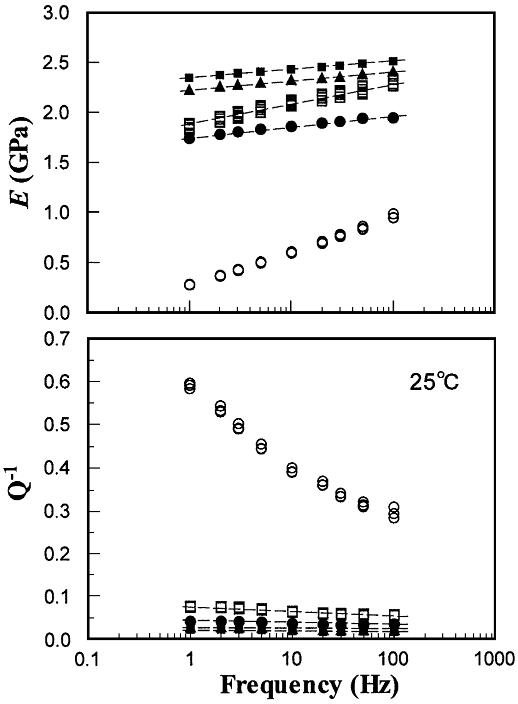

Fig. 4 Frequency variations of E and Q^{-1} at 25°C for the lacquer films. Symbols, see Fig. 2.

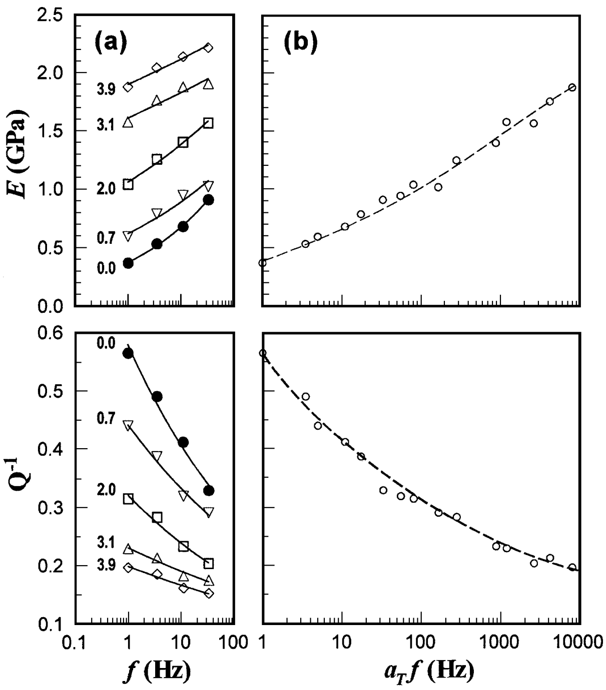

Fig. 5 Frequency variations of E and Q^{-1} for the CL lacquer film. (a), Experimental values at 25°C (●), 20°C (▽), 10°C (□), 0°C (△) and −10°C (◇) plotted against the measuring frequency (f); (b), calculated values at 25°C plotted against the reduced frequency ($a_T f$), values beside plots indicate the logarithms of the shift factor ($\log a_T$).

3.3. Mechanical Model for the Wood with Coatings

To make clear understanding of the effects of coating on the vibrational properties of wood, it is necessary to generalize the vibrational properties of coated wood with an appropriate model. Although some researchers have dealt with the effects of coating on the radiated sound [8, 9], to our knowledge, only Ono has tried to relate the vibrational properties of coated wood to those of wood and coating layer [6].

Figure 6 illustrates the most simple model for the coated wood, namely model I, proposed by Ono. This model is based on the assumption that the lacquer forms its thin layer on the surface of wood without penetration. The coated wood specimen is regarded as a composite plate consisting of the coating layer (1) and uncoated wood (0). When the t_1/t_0 is small enough to ensure $(t_1/t_0)^2 \ll 1$, the dynamic Young's modulus (E_c) and internal friction (Q_c^{-1}) of coated wood are approximately expressed by

$$E_c \approx E_0 \left[1 + 3\left(\frac{E_1}{E_0} - 1\right)\frac{t_1}{t_0}\right], \quad (1)$$

$$Q_c^{-1} \approx Q_0^{-1} \frac{1 + 3\dfrac{E_1}{E_0}\dfrac{t_1}{t_0}\dfrac{Q_1^{-1}}{Q_0^{-1}}}{1 + 3\dfrac{E_1}{E_0}\dfrac{t_1}{t_0}}, \quad (2)$$

where the suffixes 0, 1 and c indicate the uncoated wood, coating layer, and coated wood, respectively. The E_1 and Q_1^{-1} values for the coatings are given in Table 2. The t_1 value was experimentally obtained from the thickness of wood specimen before and after coating. The experimental values of E_c/E_0 and Q_c^{-1}/Q_0^{-1} are plotted against the corresponding calculated values in Figs. 7 and 8, respectively. With respect to the E_c/E_0 in the L direction and Q_c^{-1}/Q_0^{-1} in the R direction, the calculated values agreed well with the experimental ones. However, the calculated values of E_c/E_0 in the R direction and those of Q_c^{-1}/Q_0^{-1} in the L direction deviated from the experimental values. Thus, the model I seems insufficient to describe the effects of coatings on the vibrational properties of wood.

The problem is in the assumption that no lacquer penetrates into the wood. Figure 9 shows the experimental values of coating thickness (t_1) plotted against the thickness calculated from the weight and density of coatings (t_1'). The t_1 and t_1' values should be identical if no lacquer penetrates into the wood. However, the t_1 values were always smaller than the t_1' values. This fact indicates a part of lacquer penetrating into the wood did not contribute to the apparent thickness of coating. As the wood consists of porous cells, a lot of cell cavities are opened at the surface of a wood specimen.

The lacquer constituents can hardly penetrate into the wood cell wall, but easily into the "open" cavities. It seems reasonable that the difference between t_1 and t_1' was 70 μm at most, within the range of the diameter for the spruce wood cells.

Figure 10 presents a model considering the penetration of lacquer. In this model II, an additional layer (the 2nd layer) consisting of the penetrating lacquer and the wood cell wall is considered, as shown in Fig. 10(a). In microscopic level, the thickness of the 2nd layer is not constant through the specimen because of the wide variation in the diameter of natural wood cells. However, two assumptions are now introduced for the simplicity. First, the thickness of the 2nd layer is macroscopically constant. Second, the penetrating lacquer adheres tightly to the adjacent wood cell wall without any interstice. In this case, the thickness of the 2nd layer (t_2) is related to the t_1 and t_1' by

$$t_2 = \frac{t_1' - t_1}{1-\phi}, \tag{3}$$

where the ϕ is the average volume fraction of the wood cell wall in the specimen. The ϕ value can be deduced from the density of wood (ρ_0) and that of the wood cell wall ($\rho_w \approx 1.43$ [10]) by the following equation,

$$\phi = \frac{\rho_0}{\rho_w} \tag{4}$$

By modifying the Eqs. (1) and (2), the E_c and Q_c^{-1} values of coated wood are given by

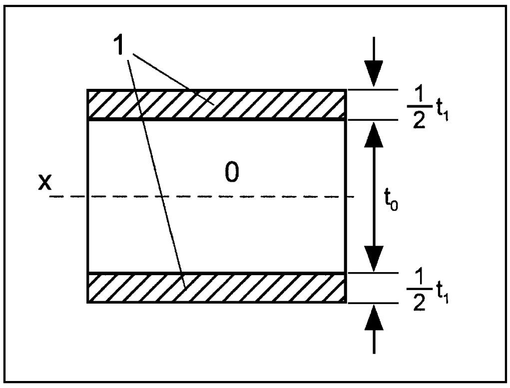

Fig. 6 The cross section of a coated wood specimen assuming no lacquer penetrates into the wood (Model I). 0, uncoated wood; 1, coating.

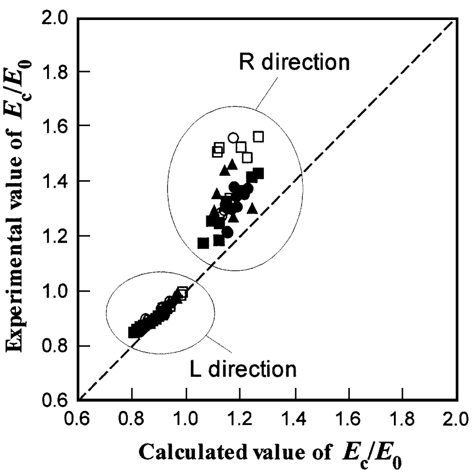

Fig. 7 Experimental values of E_c/E_0 plotted against the values calculated by using the model I. Symbols, see Fig. 2.

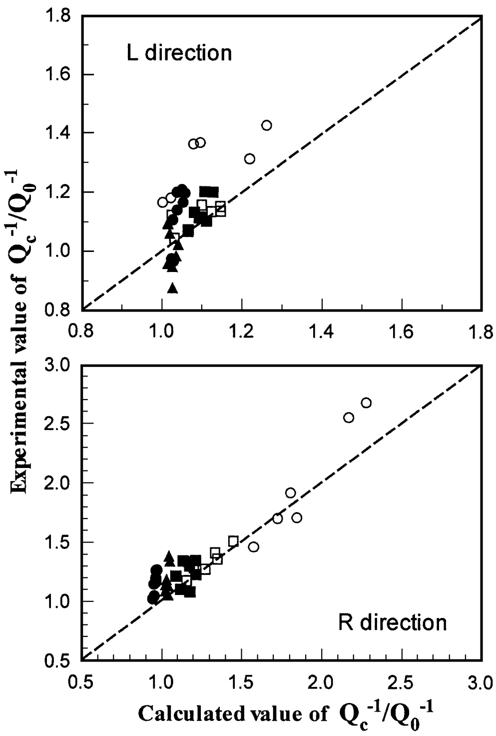

Fig. 8 Experimental values of Q_c^{-1}/Q_0^{-1} plotted against the values calculated by using the model I. Symbols, see Fig. 2.

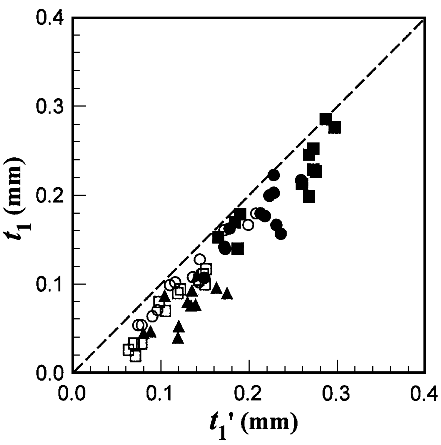

Fig. 9 Relationships between the coating thickness experimentally obtained (t_1) and those calculated by assuming no lacquer penetrates into the wood specimen (t_1'). Symbols, see Fig. 2.

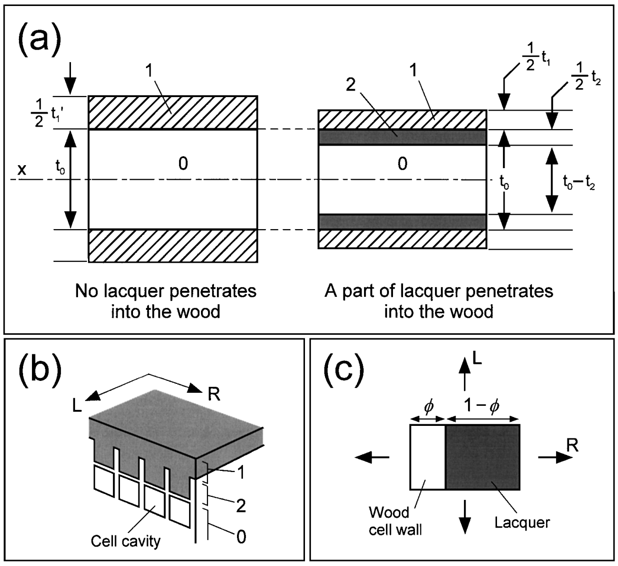

Fig. 10 A model for a coated wood considering the penetration of lacquer into the cavities of wood cells (Model II). (a), The cross section of a coated wood specimen; (b) schematic illustration at the surface of the wood specimen; (c), a model for the 2nd layer consisting of the penetrating lacquer and the wood cell wall, 0, uncoated wood; 1, coating; 2, composite layer in which the cell cavities are completely filled with lacquer.

$$E_c \approx E_{02}\left[1+3\left(\frac{E_1}{E_{02}}-1\right)\frac{t_1}{t_0}\right], \quad (5)$$

$$Q_c^{-1} \approx Q_{02}^{-1}\frac{1+3\dfrac{E_1}{E_{02}}\dfrac{t_1}{t_0}\dfrac{Q_1^{-1}}{Q_{02}^{-1}}}{1+3\dfrac{E_1}{E_{02}}\dfrac{t_1}{t_0}}, \quad (6)$$

where the E_{02} and Q_{02}^{-1} are the E and Q^{-1} values of wood including the 0th and 2nd layers, respectively. E_{02} and Q_{02}^{-1} are expressed by

$$E_{02} \approx E_0\left[1+3\left(\frac{E_2}{E_0}-1\right)\frac{t_2}{t_0-t_2}\right], \quad (7)$$

$$Q_{02}^{-1} \approx Q_0^{-1}\frac{1+3\dfrac{E_2}{E_0}\dfrac{t_2}{t_0-t_2}\dfrac{Q_2^{-1}}{Q_0^{-1}}}{1+3\dfrac{E_2}{E_0}\dfrac{t_2}{t_0-t_2}}, \quad (8)$$

where the E_2 and Q_2^{-1} are the E and Q^{-1} values of the 2nd layer, respectively.

Next, we have to derive the vibrational properties of the 2nd layer, E_2 and Q_2^{-1}, in both directions of wood. As shown in Fig. 10(b) and (c), it was assumed that the wood cell wall and penetrating lacquer in the 2nd layer are aligned in parallel along the L direction and in series along the R direction. Since the imaginary part of their complex modulus are always enough smaller than the real part, the E_2 and Q_2^{-1} in the L and R directions are expressed by,

$$E_2^{\rm L} = \phi E_{\rm w}^{\rm L} + (1-\phi)E_1, \quad (9)$$

$$Q_2^{\rm L} = \frac{\phi E_{\rm w}^{\rm L} Q_{\rm w}^{\rm L-1} + (1-\phi)E_1 Q_1^{-1}}{\phi E_{\rm w}^{\rm L} + (1-\phi)E_1}, \quad (10)$$

$$E_2^{\rm R} \approx \left(\frac{\phi}{E_{\rm w}^{\rm R}} + \frac{1-\phi}{E_1}\right)^{-1}, \quad (11)$$

$$Q_2^{\rm R-1} \approx \left[\frac{\phi Q_{\rm w}^{\rm R-1}}{E_{\rm w}^{\rm R}} + \frac{(1-\phi)Q_1^{-1}}{E_1}\right]\left(\frac{\phi}{E_{\rm w}^{\rm R}} + \frac{1-\phi}{E_1}\right)^{-1}, \quad (12)$$

where the suffixes L and R indicate the L and R directions of wood specimen, respectively, and the w indicates the wood cell wall. The $E_{\rm w}^{\rm L}$ and $Q_{\rm w}^{\rm L-1}$ values are equivalent to the experimental values of $\rho_{\rm w}E_0^{\rm L}/\rho_0$ and $Q_0^{\rm L-1}$, respectively [10]. On the other hand, the $E_{\rm w}^{\rm R}$ value was calculated from the Young's modulus of the cell wall constituents. In the thickness direction of the wood cell wall, the cellulose crystalline region and the other amorphous substances mainly consisting of lignin are aligned in series. The former and the latter have 27 GPa [11] and 4 GPa [12] as their Young's modulus, respectively. Since the cellulose content in the wood is almost 50% and its crystallinity are about 60% or higher, the volume fraction of cellulose crystalline may be in the range of 30 to 50%. Consequently, the possible value of $E_{\rm w}^{\rm R}$ are in the range of 5 to 7 GPa. In this calculation, the 6 GPa is adopted as the $E_{\rm w}^{\rm R}$ value. As there is little information about the Q^{-1} values of cell wall constituents, the $Q_{\rm w}^{\rm R-1}$ value was represented by the $Q_0^{\rm R-1}$ value determined experimentally. Finally, the E_c/E_0 and Q_c^{-1}/Q_0^{-1} values can be deduced from the local quantities by combining Eqs. (3) to (12). These values are compared to the experimental values in Figs. 11 and 12, respectively. Considering the simplicity of model and the natural variation in the wood cells, the agreement with the experimental data was fairly good. This fact suggests that the penetrating lacquer should be taken into consideration when we want to predict the effects of coatings on the vibrational properties of wooden-soundboards.

4. CONCLUSIONS

The effects of urushi coatings on the vibrational properties of wood were compared to those of some conventional lacquer coatings. The urushi was similar to the polyurethane lacquer for the soundboard of harp in viscoelastic and mechanical properties. These results suggested the possibility of urushi as a coating for the harp soundboard. The effects of coatings on the vibrational properties of wood were expressed by a model consisting of three layers, wood, coating, and composite layer of the penetrating lacquer and the wood cell wall.

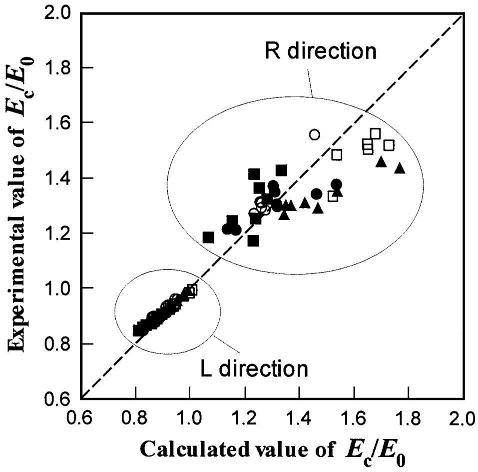

Fig. 11 Experimental values of E_c/E_0 plotted against the values calculated by using the model II. Symbols, see Fig. 2.

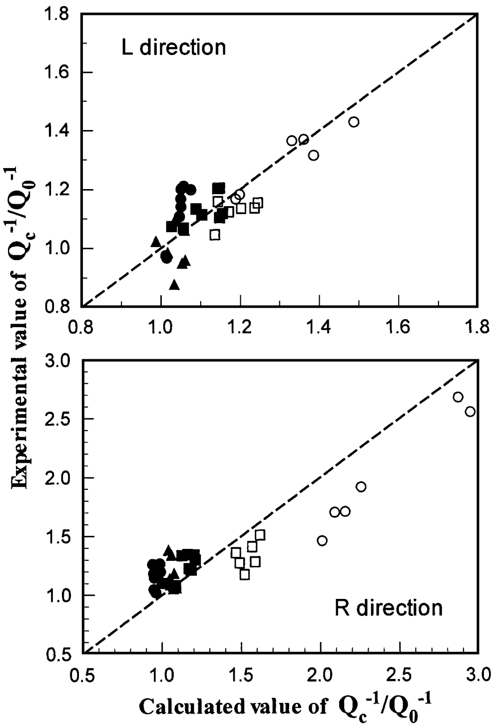

Fig. 12 Experimental values of Q_c^{-1}/Q_0^{-1} plotted against the values calculated by using the model II. Symbols, see Fig. 2.

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