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Stress relaxation of wood during elevating and lowering processes of temperature and the set after relaxation II: consideration of the mechanism and simulation of stress relaxation behavior using a viscoelastic model

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Abstract Previously we showed that the relaxation modulus *Et* of water-saturated wood during temperature reduction maintained its initial value despite the decrease in temperature, although during temperature elevation *Et* showed a marked decrease. In the present study, to clarify the mechanism of relaxation during temperature elevation and reduction, Young's modulus was measured in stress relaxation experiments with changes in temperature, and relaxation behavior was simulated using a Maxwell model consisting of five elements. Furthermore, the dynamic Young's modulus and dynamic loss modulus were measured during both temperature elevation and reduction. The results obtained suggested that the unique relaxation behavior during temperature reduction was caused by decreases in Young's modulus and coefficient of viscosity (i.e., an increase in fluidity) compared with those during elevation of temperature. The decrease in Young's modulus and increase in fluidity were considered to be due to an unstable structure in wood that occurred during temperature reduction. This unstable structure probably develops in the nonequilibrium state of temperature toward a true equilibrium state. Wood should be more unstable during temperature reduction than during temperature elevation because of the decrease in molecular motion when the temperature is lowered.

Key words Wood · Stress relaxation · Elevating temperature · Lowering temperature · Viscoelastic model

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Introduction

In our previous study[1] we showed that the stress relaxation of water-saturated wood during temperature elevation was almost equal to that at the higher constant temperature within the range of temperature elevation. This result differed from those of previous reports. Moreover, although the relaxation modulus *Et* during temperature elevation decreased markedly with increasing temperature, during temperature reduction it remained constant despite the decreased temperature. The reason for the latter observation is not clear. It is important to clarify the mechanism to understand the fundamental viscoelastic properties of wood during temperature elevation and reduction. In addition, the set after stress relaxation occurred to a greater extent during the temperature-lowering process than during temperature elevation. However, the mechanism underlying this phenomenon has yet to be determined. In all previous studies[2–4] creep deformation was measured and discussed. Hence, it is necessary to investigate the relation between the behavior of stress relaxation and creep deformation during both temperature elevation and reduction.

The purpose of the present study was to determine the mechanism of stress relaxation during temperature elevation and reduction. Young's modulus was measured in stress relaxation experiments during elevation and reduction of temperature; then relaxation behavior was simulated using a Maxwell model consisting of five elements that have physical meaning. Furthermore, the dynamic Young's modulus and dynamic loss modulus during both processes (temperature elevation and reduction) were measured to verify the simulated results.

Materials and methods

Test specimens

Wood samples were obtained from the heartwood of a hinoki log (*Chamaecyparis obtusa*). To measure the relaxation modulus, the elastic modulus of wood during stress relaxation and creep deformation (limited measurement), more than 100 radial sectional wood pieces [12 (radial, R) × 1 (tangential, T) × 0.4 (longitudinal, L) cm] were successively cut from several wood sticks with a cross section of 12 (R) × 1 (T) cm and a length of about 40 cm. To measure the dynamic elastic modulus and loss modulus in vibrational tension, more than 20 radial sectional wood pieces [3 (R) × 0.3 (T) × 0.15 (L) cm] were successively cut from a wood stick with a cross section of 3 (R) × 0.3 (T) cm. These specimens were oven-dried and then saturated with water under vacuum.

Measurement of stress relaxation and Young's modulus during stress relaxation under conditions of temperature elevation and reduction

The apparatus used for stress relaxation and Young's modulus during bending in the stress relaxation experiment was similar to that described in our previous paper.[1] Test samples were subjected to prescribed initial deflections by moving upward a load cell installed on the axis of a mobile system; the deflections were measured with a laser displacement meter. The loads were measured by the load cell, and the rates of temperature elevation and reduction were controlled by a temperature controller (model SU type, Chino Corporation). Test samples were supported at two points, and the initial deflections were applied to the center of the specimen with a span of 8 cm. Moduli of elasticity were measured by sudden addition and immediate removal of a small deflection at several temperatures during the stress relaxation experiment.

Six temperatures (20°C, 40°C, 50°C, 60°C, 70°C, 80°C) were chosen for the measurement of stress relaxation at constant temperature. The stress relaxation during both temperature elevation and reduction was measured under five conditions: 20°–80°C, 20°–60°C, 20°–40°C, 40°–60°C, 60°–80°C. Initial deflections were mainly 1, 2, and 3 mm for limited numbers of samples. The stress relaxation was measured for 10 h. For the initial 4 h relaxation was measured at an initial constant temperature, during the next 4 h at a constant rate of temperature elevation or reduction, and for the final 2 h at the final temperature.

Measurement of creep deformation, dynamic Young's modulus, and dynamic loss modulus of wood during temperature elevation and reduction

Creep deformation during bending was measured using apparatus and methods similar to those described for the stress relaxation measurement. The dimension of the specimens, temperature control, support of the specimen, and the span were the same as those described for the stress relaxation experiment. A constant load of 100 g was applied. The amount of deflection was measured with a linear transducer during temperature elevation from 20°C to 80°C and during temperature reduction from 80°C to 20°C. For the initial 4 h the temperature was controlled at the initial temperature, for the next 4 h there was a constant rate of temperature elevation or reduction, and for the final 2 h the temperature was kept at the final temperature, as for the stress relaxation measurement.

The dynamic Young's modulus (E') and dynamic loss modulus E'' were measured during vibrational tension (0.05 Hz, 100–200 Pa) at several temperatures during temperature elevation and reduction using a thermomechanical analyzer (TMA-6100 type, Seico Instruments). Two kinds of wood sample (i.e., green wood and water-saturated wood) were chosen for the measurement of E' and E'' to analyze the stress relaxation mechanism during temperature elevation and reduction.

Results and discussion

Stress relaxation and creep deformation during temperature elevation and reduction

Figure 1 shows stress relaxation curves for relations between the relaxation modulus *Et* and temperature during temperature elevation from 20°C to 80°C and temperature reduction from 80°C to 20°C. Figure 2 shows creep compliance curves under the same conditions as for stress relaxation. These curves differed markedly in the amounts of stress relaxation and creep compliance for temperature elevation and reduction. However, the decrease in *Et* during temperature elevation corresponded to the increase in creep compliance during temperature elevation. Values for *Et* and creep compliance remained almost constant during temperature reduction. Thus, the relation between *Et* and creep compliance during temperature elevation and reduction can be regarded as a mirror image of the temperature axis. Consequently, we can discuss the viscoelastic properties of wood

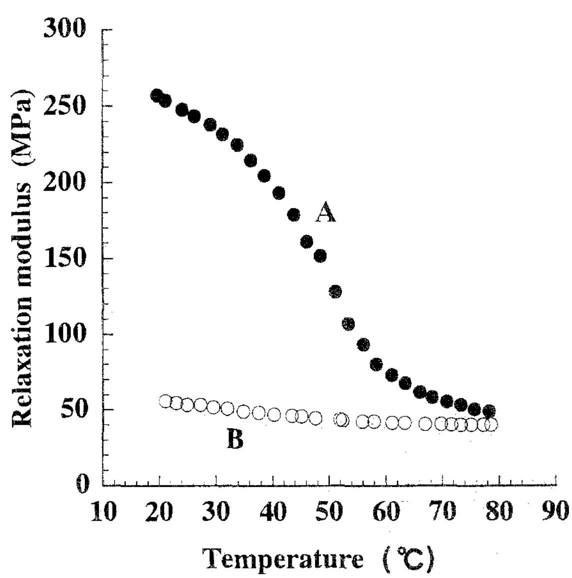


Fig. 1. Stress relaxation of wood during temperature elevation and reduction shown as the relation between relaxation modulus and temperature. *A*, elevation of temperature; *B*, reduction of temperature

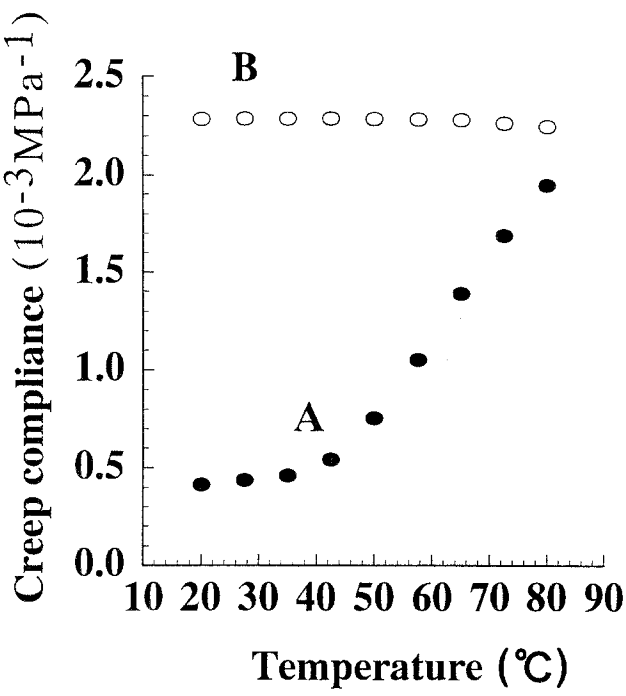


Fig. 2. Changes in creep compliance during the elevation and reduction of temperature. *A*, elevation of temperature; *B*, reduction of temperature

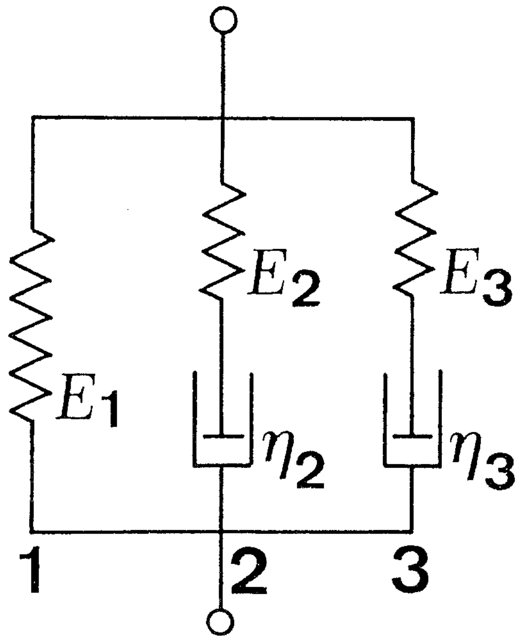


Fig. 3. Viscoelastic model of wood for simulation. E_1, E_2, E_3, moduli of elasticity; η_2, η_3, coefficients of viscosity. E_2 and η_2 are related to moisture content of wood; E_3 and η_3 vary with temperature

during temperature elevation and reduction based on either stress relaxation or creep deformation behavior.

Viscoelastic model and simulations using the model

Figure 3 shows a viscoelastic model[5] of wood for simulating stress relaxation behavior during temperature elevation and reduction. The Maxwell model consists of five elements: E_1, E_2, and E_3 are moduli of elasticity, and η_2 and η_3 are coefficients of viscosity.

Each element has physical meaning as follows: an isolated elastic element in *1* in Fig. 3 was necessary because stress never relaxed to zero, elastic and viscous elements in *2* (Fig. 3) are related to moisture content of wood, and elastic and viscous elements in *3* (Fig. 3) were sensitive to temperature.

According to the viscoelastic theory,[6] the relaxation modulus *Et* of the proposed model is represented by the following equation.

$$E_t = E_1 + E_2\left(\frac{1}{e^{\frac{E_2}{\eta_2}t}}\right) + E_3\left(\frac{1}{e^{\frac{E_3}{\eta_3}t}}\right)$$

where t is time.

Appropriate values of moduli of elasticity and coefficients of viscosity of each element were applied so the simulated result for temperature elevation coincided with the stress relaxation behavior measured during the process of temperature elevation, and the relaxation behavior during the process of temperature reduction was also simulated using the same values at the same temperatures as those used for elevating temperatures.

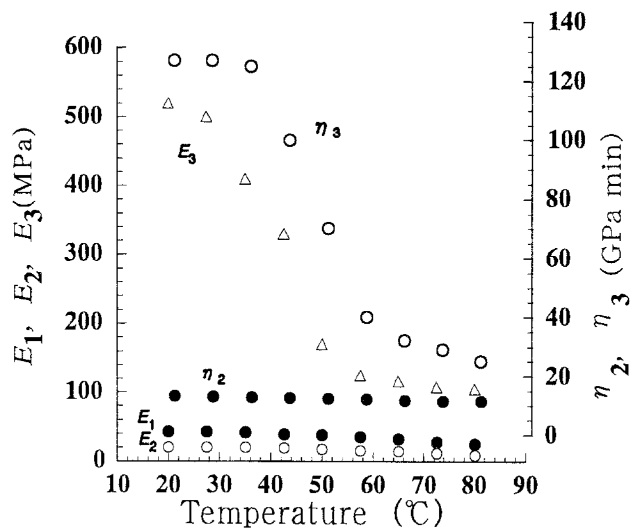


Fig. 4. Values of E_1, E_2, E_3, η_2, and η_3 for simulations. Values were the same at the same temperatures for both processes of temperature elevation and reduction

Figure 4 shows the values applied for each element. The simulated values of *Et* are shown in Fig. 5 for a comparison with the measured values of relaxation modulus *Et* during temperature elevation and reduction. As shown in Fig. 5, simulated values for temperature reduction did not agree with the measured values over the whole temperature range; that is, the simulated values were always larger than the measured values. This result suggested that different values in each element should be applied for temperature elevation and reduction.

It is not known for which processes of temperature elevation or reduction larger elastic moduli should be applied. Young's moduli of wood samples during both temperature elevation and reduction were measured by sudden application and immediate removal of a small deflection during the stress relaxation measurements. The results are shown in Fig. 6. Elastic moduli during temperature reduction were smaller than those while elevating the temperatures at temperatures lower than 50°C. This result means that a smaller elastic modulus should be applied to E_3 during temperature reduction than during elevation. We then examined appropriate values of η_3 consistent with the measured E_t by simulation using lower E_3 for temperature reduction. The result indicated that lower η_3 had to be applied for temperature reduction than for temperature elevation. The applied values for each element are shown in Fig. 7, and the results of the simulation are shown in Fig. 8.

As shown in Fig. 8, the simulated values agreed with the measured values during both processes. Accordingly, it was clear that E_3 and η_3 during temperature reduction were smaller than those during temperature elevation. This was thought to indicate that during temperature reduction wood not only has lower elasticity but also higher fluidity than during temperature elevation.

Figure 9 shows the dynamic Young's modulus E' and dynamic loss modulus E'' measured during temperature elevation and reduction. E' had smaller values during the process of temperature reduction than during elevation, consistent with the results shown in Fig. 6. On the other hand, E'' values during temperature reduction were larger than those during temperature elevation. Comparing E'' values with the η_3 values shown in Fig. 7, and judging from the fact that an increase in E'' of water-saturated wood were found to be caused by decreases in η_3,[5] the behavior of E'' shown in Fig. 9 was consistent with that of η_3 shown in Fig. 7. Based on these results, we concluded that wood during temperature reduction shows lower elasticity and higher fluidity than wood during temperature elevation. The mechanism of this phenomenon is discussed in the following section.

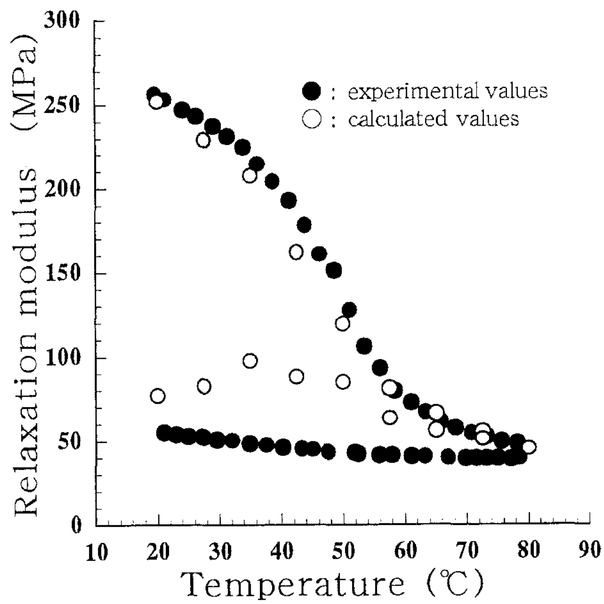


Fig. 5. Comparison of experimental and simulated values when the elements were given the values shown in Fig. 4

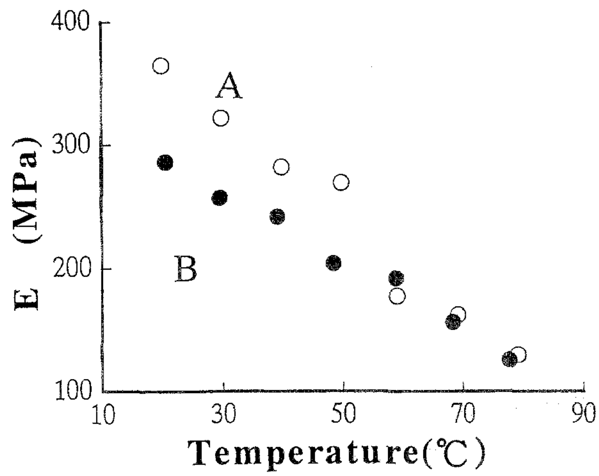


Fig. 6. Temperature dependence of Young's modulus E during bending. A, elevation of temperature; B, reduction of temperature

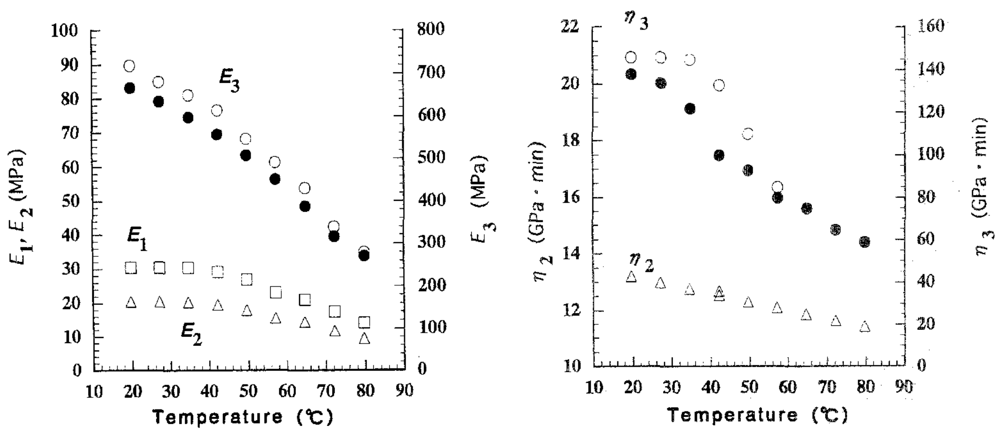


Fig. 7. Values given to each element for the simulation. E_3 and η_3 were given different values for the processes of temperature elevation (*open symbols*) and reduction (*filled symbols*)

Mechanism of the differences in viscoelastic properties of wood for temperature elevation and reduction

As mentioned in the preceding section, it is clear that wood has lower elasticity and higher fluidity during temperature reduction than during elevation. Thus, it is necessary to consider what types of structural change in wood are responsible for these differences in viscoelastic properties.

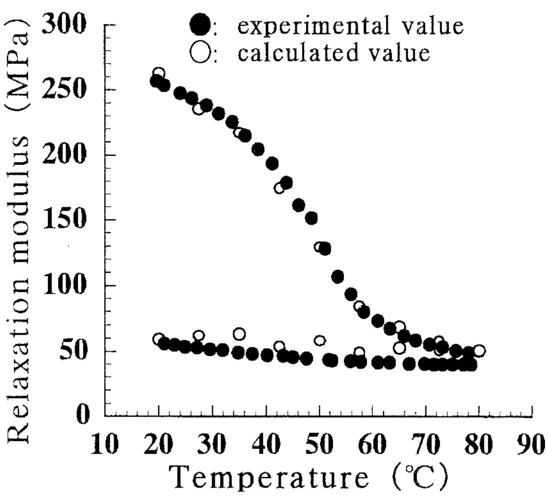


Fig. 8. Comparison of experimental and simulated values when the elements were assigned the values shown in Fig. 7

The decrease in elasticity and increase in fluidity are probably caused by a similar unstable state in the internal structure of wood as suggested by Ishimaru et al.,[7,8] who investigated the changes in the physical and mechanical properties during and after a period of moisture conditioning. They found that the modulus of elasticity and modulus of rupture during the bending of wood conditioned by adsorption of moisture showed a significant increase during the later stage of conditioning when moisture content scarcely changed, and that stress relaxation decreased with increases in the conditioning period during both adsorption and desorption. These results suggested that wood in an unstable state caused by the existing state of moisture different from that in a true equilibrium state; it has lower elasticity and strength and higher fluidity than wood in a true equilibrium state. This unstable state was interpreted as a phenomenon that occurs during stabilizing processes, such as the reorientation of molecular chains of wood constituents toward a true swelling equilibrium by adsorption of moisture. Also, such an unstable state was shown to affect fluidity more extensively than the elasticity or strength of wood. These characteristics were consistent with the viscoelastic properties determined in the present study

Fig. 9. Changes in E' and E'' during the processes of temperature elevation and reduction. E', dynamic elastic modulus; E'', dynamic loss modulus; *A*, elevation of temperature; *B*, reduction of temperature

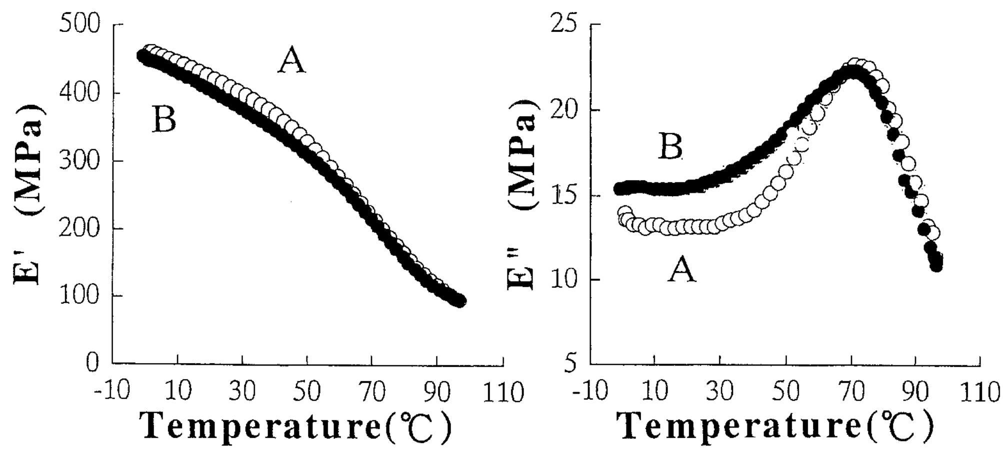
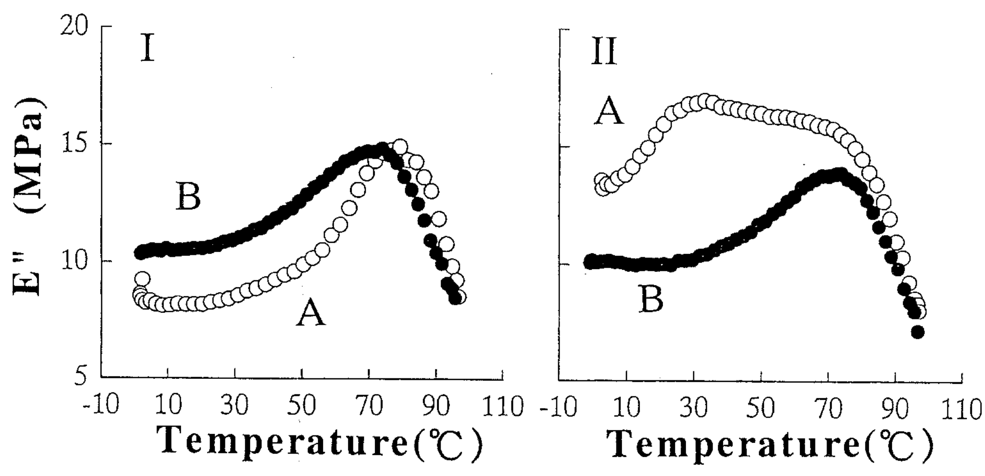


Fig. 10. Changes in E'' for green wood **(left)** and water-saturated wood after drying once **(right)** during elevation and reduction of temperature. *A*, elevation of temperature; *B*, reduction of temperature



that showed decreases in the elastic modulus and coefficient of viscosity caused by cooling.

Furuta et al.[9] reported that a green specimen had the largest elastic modulus, and the specimen resaturated with water after drying and the water-saturated specimen rapidly cooled after heating showed markedly decreased elastic moduli. They attributed these results to the release of the strain produced by drying and rapid cooling. Ishimaru et al.[10] found that wood swollen in organic liquids distant from the swelling equilibrium exhibited lower elasticity and strength than expected from their degree of swelling, that wood swollen in liquids during the second process of temperature elevation showed lower E' than during the first temperature elevation, and the thermal softening found during the first temperature elevation disappeared during the second increase in temperature. Based on these results, they concluded that wood in an unstable state distant from the swelling equilibrium was highly flexible and weak.

The relaxation behavior in the present study, especially that during temperature reduction, can be interpreted as resulting from an unstable state induced by rapid cooling after heating. The equilibrium state of wood should vary with temperature. Therefore, when temperature changes, the wood is no longer in the equilibrium state. Under such conditions, some strain should occur in the cell wall structure, probably based on changes in the distribution of moisture in the cell wall with temperature or a difference in thermal expansion among wood constituents (or both). The unstable state of wood mentioned above can be interpreted as a state with internal strain. Furthermore, such strain is considered to be more easily released during temperature elevation than during reduction because molecular motions of moisture and the wood constituents are increasingly activated with increasing temperature.

To ascertain the validity of the above interpretation, the changes in E'' during temperature elevation and reduction were measured using green wood and water-saturated wood after drying. Green wood, which is thought to be in the most stable state, was predicted to show larger E'' during temperature reduction than during temperature elevation because of the unstable structure produced by lowering of temperature. On the other hand, the resaturated wood, which is thought to be in a highly unstable state due to drying, was predicted to have a smaller E'' during temperature reduction than during elevation because the highly unstable structure produced by drying should be modified with the increase in temperature based on activation of molecular motion. Therefore, E'' is considered to be less unstable during temperature reduction than during temperature elevation.

Figure 10 shows the differences in E'' in the wood specimens for temperature elevation and reduction. Green wood, while lowering the temperature, has a larger E'' at relatively lower temperature ranges than while elevating the temperature. On the other hand, water-saturated wood after being dried once showed an opposite result. These results agreed with the predictions. Accordingly, the interpretation and consideration mentioned above can be regard as valid.

Based on these results, the relaxation behavior of wood during both temperature elevation and reduction (i.e., E_t while elevating the temperature) decreased with the changing temperature; however, E_t while lowering the temperature remained almost constant despite the temperature reduction. This can reasonably be explained by a decrease in elastic modulus and an increase in fluidity resulting from an unstable structure in the cell wall produced during the process of lowering the temperature. Furthermore, the unstable state was thought to have resulted from some internal strain probably induced by changes in the existing state of moisture with temperature or differences in thermal expansion among wood constituents (or both).

Conclusions

The causes of the differences between stress relaxation behavior during temperature elevation and reduction were discussed using the Maxwell model, which consists of five elements. Young's moduli were measured during the course of relaxation measurements with both elevation and reduction of temperature by immediately adding and removing a small deflection. The dynamic Young's modulus and dynamic loss moduli during vibrational stretching were also measured during both temperature elevation and reduction.

Simulation using the Maxell model with the same elastic moduli and coefficients of viscosity for each element of the model was not consistent with the measured results obtained during elevation or reduction of temperature. Young's moduli measured during the process of temperature reduction were smaller than those during the process of temperature elevation. Hence, relaxation behavior was again simulated by assuming lower elasticity and larger fluidity for the process of temperature reduction. As a result, the simulated values showed good agreement with the experimental values throughout temperature elevation and reduction.

Based on these results, the relaxation behavior of wood during temperature elevation and reduction (i.e., the relaxation modulus while elevating the temperature decreased with the rise in temperature, although the relaxation modulus while lowering the temperature remained almost constant despite the temperature reduction) can reasonably be explained by a decrease in elastic modulus and an increase in fluidity due to an unstable structure in the cell wall produced during temperature reduction.

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