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ORIGINAL ARTICLE

Temperature dependence of the dynamic viscoelasticity of bases of Japanese cypress branches and the trunk close to the branches saturated with water

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Abstract With the aim of obtaining findings on the dynamic properties of branches and their bases, as well as their support mechanisms, the present study examined the temperature dependence of the dynamic viscoelasticity of Japanese cypress samples saturated with water to clarify the responses in different regions, and identified factors influencing the characteristics. In the bases of the branches: E' sharply decreased at approximately room temperature and significantly decreased at around 20 and 60 °C; a peak and shoulder peak of E'' or tan δ were noted at around 20 °C, and there was another peak of tan δ at around 60–80 °C; and mechanical relaxation was noted at around 20 °C and 60–80 °C. On the other hand, in some regions, including the trunks, branches, and their bases, mechanical relaxation was only noted on the high-temperature side. However, boiling treatment with about 12 % weight loss inhibited mechanical relaxation, and there were decreases in E', E'', and tan δ at approximately room temperature. The bases of the branches of Japanese cypress are considered to develop its elasticity and viscosity to tolerate external stress by accumulating an extract, which enhances the strength of lignin.

Keywords Japanese cypress · Bases of branches · Water-saturated condition · Dynamic viscoelasticity · Mechanical relaxation

Introduction

It is difficult to use branches and their bases for wood-based materials. As a disadvantage of the bases of branches, wood products using them are not strong, and it is difficult to compress or process them. It is difficult to utilize the branches because they are thinner than the trunks and parts are often reaction wood. However, to promote the effective use of wood, it is important to acquire detailed knowledge of these regions and discuss the methods for their utilization.

Branches are essentials for trees to grow. The branches of a tree serve to support its leaves, which are necessary for growth. They usually grow horizontally against gravity to become exposed to as much light as possible. Therefore, the branches are more easily affected by rain, winds, and other external forces, as compared to the trunks, and large stress is applied to the bases of the branches in particular. Furthermore, trees in some areas are influenced by large temperature changes, in addition to strong winds and heavy rain. This means that the branches are supported by their bases under very severe conditions. Therefore, studies on various aspects of the dynamic properties of branches and their bases are essential to understand the support mechanisms. Moreover, these studies are also very significant from the viewpoint of biometric research because trees have acquired their attributes over the course of evolution, which may be applied for the development of new materials. Although there are some studies on the dynamic properties of branches and their bases focusing on the

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growth stress and elastic modulus at approximately room temperature, no studies have been conducted to examine the viscoelasticity of them at varying temperatures.

We have already obtained a number of findings on the dynamic viscoelasticity of wood saturated with water. One of these findings is that non-crystalline sugars, including hemicelluloses whose moisture content level is the same as or higher than the fiber saturation point, soften at approximately −40 °C; lignin softens at approximately 80 °C [1]. Our previous studies also suggest that wood whose moisture content is approximately 20 % or higher and non-crystalline sugars contained in moso bamboo (*Phyllostachys edulis*) soften at approximately −40 °C, lignin softens at approximately 80 °C, and the softening temperature of lignin varies between softwood, Japanese hardwood, and tropical hardwood [2–6]. The study results also suggest that the dynamic viscoelasticity of wood saturated with water is significantly influenced by the history of drying, heat history in the water (cooling methods, etc.), and immersion time, and that the temperature causing the greatest loss of support attributed to the softening of lignin varies depending on the direction of fibers and their orthogonal direction [7–11].

With the aim of obtaining findings on the dynamic properties of branches and their bases as well as the support mechanisms of the trees, the present study examined the temperature dependence of the dynamic viscoelasticity of Japanese cypress samples, collected from the trunks, branches, and their bases, to understand their responses, based on our previous findings on the dynamic viscoelasticity of wood saturated with water. An examination was then conducted to identify factors influencing the responses, and the causes of the phenomena were discussed.

Experimental

Samples

Samples were made from a 90-year-old Japanese cypress tree (*Chamaecyparis obtusa*). The diameter at breast height of the tree was about 45 cm. Figure 1 shows images of sampled regions. Samples collected from the sapwood and heartwood of the trunks and branches, bases of the branches [(1)–(3) in Fig. 1], and parts of the trunk close to the branches [(4)–(6) in the Fig. 1] were used in experiment. The specific gravities in each region were as follows: 0.41 in the sapwood of the trunk, 0.48 in the heartwood of the trunk, 0.84 in the sapwood of the branches, 1.15 in the heartwood of the branches (1) 1.10, (2) 1.10, and (3) 0.99 in the bases of the branches, and (4) 0.60, (5) 0.50, and (6) 0.45 in parts of the trunk close to the branches. The size of specimens was 1.2 mm (*L*) × 25 to 30 mm (*R*) × 1 mm (*T*). The samples for the measurement of dynamic viscoelasticity were provided in green condition.

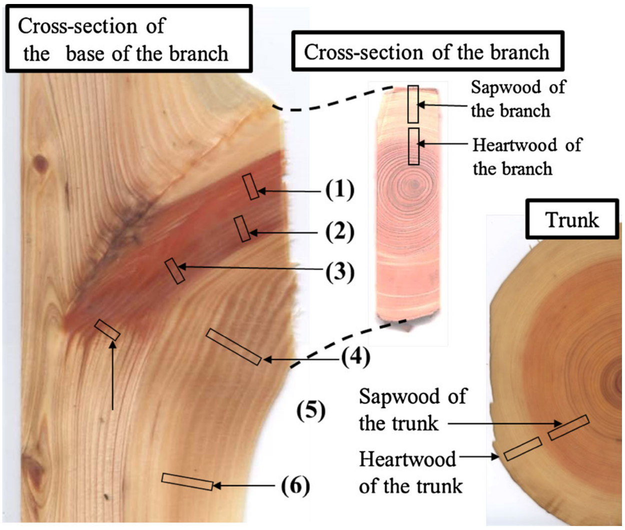

Fig. 1 Details of sampled regions

Following measurement, the samples underwent 2-h boiling extraction or methanol-based extraction treatment using Soxhlet reflux equipment (at 64 °C and for 6 h), and then they were measured again in water-saturated condition.

Measurement of dynamic viscoelasticity

Forced-oscillation-type equipment for the measurement of viscoelasticity (DMS6100 by SII Nano-Technology) was used to measure the dynamic viscoelasticity of samples. Measurement was conducted in water, while its temperature was rising: the range of temperature measurement was between 5 and 95 °C; measurement frequencies were 0.05, 0.5, 1, 5, 10, and 20 Hz; the rate of temperature change was 1 °C/min; and the measurement span was 10 mm. The tensile direction was in the radial direction. Temperature measurement was conducted during the courses of the first and second temperature rises according to the conditions. Measurement was conducted during the courses of both the first and second temperature rises, with the aim of assessing the effects of releasing the growth stress [12, 13] and degeneration of structural components [3] due to the heat history, and accurately determining the responses of each sample by ensuring that there were no differences in the histories of heat applied to them; the results of previous studies [7–9] suggest that temperature dependence of the dynamic viscoelasticity significantly varies depending on the heat history of the sample.

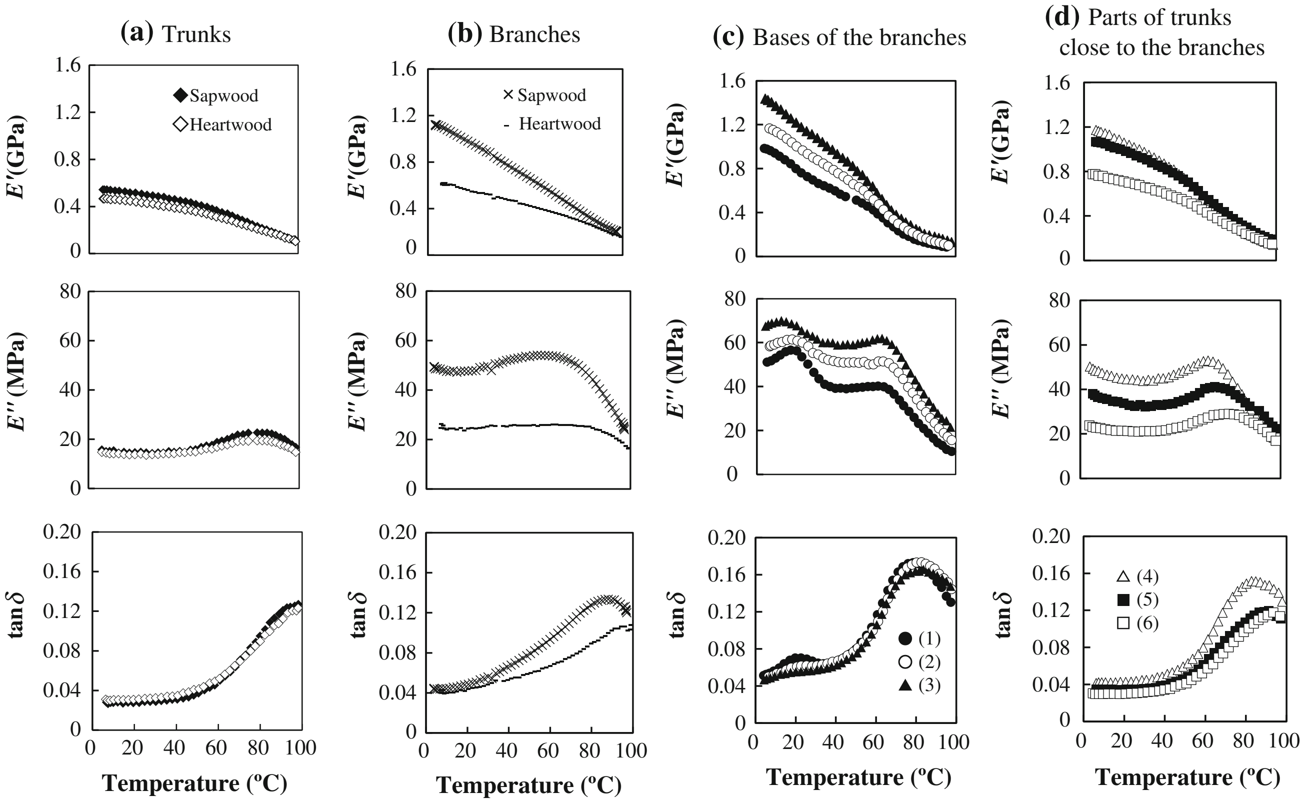

Fig. 2 Temperature dependence of the dynamic viscoelasticity of Japanese cypress sapwood and heartwood of the trunks **a** and branches **b**, bases of the branches **c**, and parts of the trunk close to the branches **d** saturated with water at 0.5 Hz. Details of sampled regions are shown in Fig. 1

Results

Figure 2 shows the storage elastic modulus (E'), loss of the elastic modulus (E''), and loss of the tan δ of the sapwood and heartwood of the trunks (Fig. 2a) and branches (Fig. 2b), bases of the branches [(1)–(3) in Figs. 1, 2c), and parts of the trunk close to the branches [(4)–(6) in Figs. 1, 2d] during the course of the first temperature rise at 0.5 Hz.

In the sapwood and heartwood of the trunk, E' sharply decreased from around 60 °C, E'' peaked at approximately 80 °C, and tan δ was markedly high at approximately 100 °C, all of which were consistent with the results of previous studies (Fig. 2a). The results in the sapwood of the branches were also similar (Fig. 2b). According to the previous studies, these peaks are attributed to the micro-Brownian motion of lignin [2–6]. In the present study, this dynamic relaxation process is defined as the α relaxation process. On the other hand, in the heartwood of the branches and parts of the trunk close to the branches [(4)–(6) in Fig. 1], E' sharply decreased from around room temperature, E'' peaked at approximately 60 °C, and tan δ peaked at approximately 80–90 °C (Fig. 2d). These results are similar to the α relaxation process that occurred in the sapwood and heartwood of the trunk (Fig. 2a) and sapwood of the branches (Fig. 2b) that shifted to the low-temperature side. In the sapwood of the branches, changes in E', E'', and tan δ were significant within the range of temperature measurements (Fig. 2b). On the other hand, in the bases of the branches [(1)–(3) in Fig. 1], there was a sharp decrease in E' at around room temperature and significant decreases at approximately 20 and 60 °C, E'' peaked at approximately 20 and 60 °C, a peak and shoulder peak of tan δ were noted at around 20 °C, and there was another peak of tan δ at around 80 °C (Fig. 2d). In the present study, this dynamic relaxation process on the low-temperature side is defined as the β relaxation process. For all of the results, differences in E' and E'' may be attributable to differences in their specific gravities. On the other hand, differences in the responses in the bases of the branches and other regions, and the occurrence of β relaxation in particular, may be due to the effects of the release of growth stress, extracted constituents, and other influences. Since the characteristics in the bases of the branches were different from those in other regions, including the occurrence of β relaxation process in particular, changes in E', E'', and tan δ in the bases of the branches during the course of the second temperature rise were also examined to identify the causes.

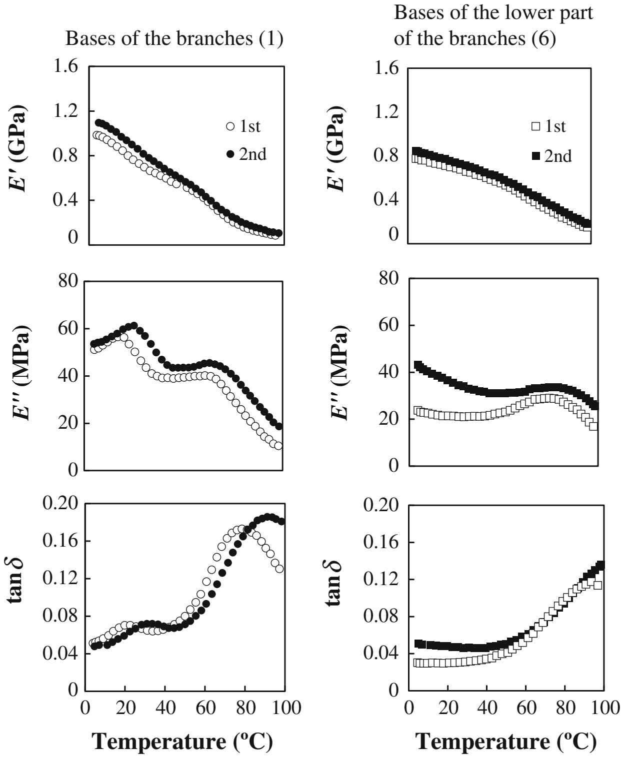

Fig. 3 Temperature dependence of the dynamic viscoelasticity of the sample (1) and (6) during the courses of the first and second temperature rise at 0.5 Hz. Details of sampled regions are shown in Fig. 1

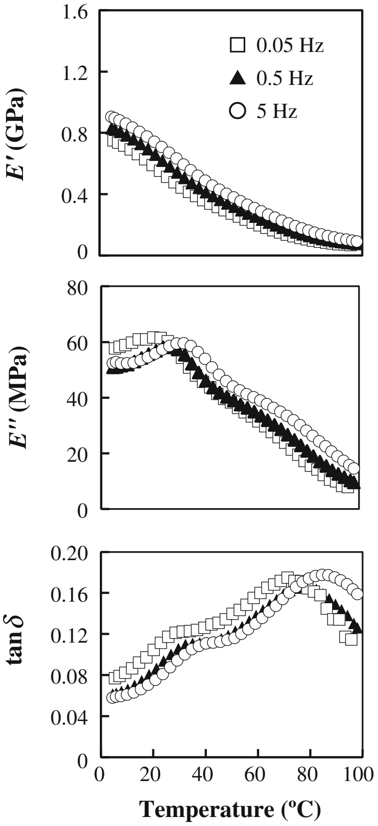

Fig. 4 Temperature dependence of the dynamic viscoelasticity of the sample (1) during the course of the second temperature rise at 0.05, 0.5, and 5 Hz. Sample No. shown in Fig. 1

Figure 3 shows E', E'', and tan δ of Sample (1) in the bases of the branches and Sample (6) collected from a part of the trunk close to the branches (a sample from a region other than the bases of the branches) during the courses of the first and second temperature rise at 0.5 Hz.

In the bases of the branches, α and β relaxation processes occurred in each of E', E'', and tan δ, which are consistent with the results during the courses of the first temperature rise. However, the range of temperatures slightly shifted to the high-temperature side. Similar results were obtained when samples in other bases of the branches were used, although they are not presented in the figures. On the other hand, those changes were not noted in the part of the trunk close to the branches, and only the α relaxation process occurred. Similar changes were also noted in all samples other than the bases of the branches, although they are not presented in the figures. These results suggest that the β relaxation process noted in the bases of the branches was not affected by the first rise temperature; in other words, this specific characteristic does not significantly change when the temperature increases by the first rise temperature. The following figure shows E', E'', and tan δ in the bases of the branches measured at different frequencies with the aim of further discussing these unique responses including the β relaxation process:

Figure 4 shows E', E'', and tan δ of Sample (1) in the bases of the branches during the course of the second temperature rise at 0.05, 0.5, and 5 Hz.

Both α and β relaxation processes were frequency dependent. The results of a previous study [5] suggest that the α relaxation process is clearly frequency dependent. On the other hand, the above-mentioned result that the β relaxation process is frequency dependent suggests that it is a type of dynamic relaxation process, such as molecular motions, which are not solely dependent on the temperature. According to the general rheological theory of polymer materials [14], the reason is as described below. Reactions such as molecular motions are dependent on temperature and time. Therefore, the reactions are dependent on the frequency and appear as dynamic relaxation process. On the other hand, reactions that depend only on temperature mainly, such as melting and crystallization, are not almost depend on the frequency. The apparent activation energy of the β relaxation process is calculated by an Arrhenius plot using the reciprocals of the peak temperatures of E'' and tan δ and the logarithms of frequencies

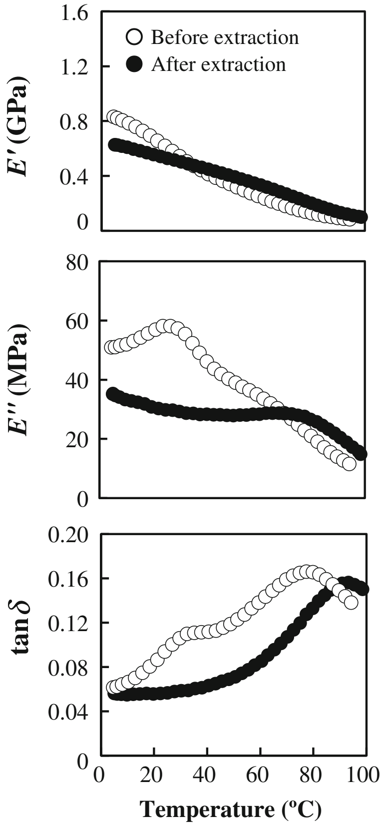

Fig. 5 Temperature dependence of the dynamic viscoelasticity of the sample (1) before and after 2-h boiling extraction treatment during the course of the second temperature rise at 0.5 Hz. Sample No. shown in Fig. 1

were approximately 350 kJ/mol. With the range of temperatures at which the β relaxation process occurs and the large amount of activation energy, the cause of the β relaxation process may be the micro-Brownian motion of molecular chains contained in the non-crystalline regions, based on the general rheological theory [15]. On the other hand, according to the results of previous studies, most non-crystalline sugars soften in the range of temperatures below zero [1, 2]. Therefore, the β relaxation process is reasonably assumed to be caused by the micro-Brownian motion of part of lignin plasticized by the extracted constituent. On the other hand, the β relaxation process during the course of the second temperature rise shifted toward the high-temperature side by approximately 10 °C, as shown in Fig. 3. This suggests that the changes are affected by the heat history. To confirm this, following the first measurement, the samples underwent 2-h boiling extraction or methanol-based extraction treatment, and then they were measured again. Samples (1)–(3) underwent these processes of extraction treatment, and the mean weight reduction rate was approximately 12 %.

As an example of the results obtained, Fig. 5 shows E', E'', and tan δ of Sample (1) in the bases of the branches before and after 2-h boiling extraction treatment during the course of the second temperature rise at 0.5 Hz.

When extraction treatment was conducted, the β relaxation process did not occur, and unique responses identified in the bases of the branches were not noted. Extraction treatment also caused a reduction in E' at approximately room temperature and significant reductions in E'' and tan δ. Similar results were obtained when samples in other bases of the branches were used or methanol extraction treatment was conducted, although they are not presented in the figures. These results suggest that the β relaxation process is a type of dynamic relaxation process that occurs when the component can be extracted, and that the component significantly increases the elasticity, and viscosity in particular, of the wood at approximately room temperature.

Discussion

It is known that large stress develops in parts of trees that grow on slopes. Since these trees attempt to maintain straight trunks, reaction wood has a different tissue and chemical structure from that of normal wood [16]. In the case of hardwood, tension wood is generated on the side of the trees facing the mountain. Tension wood produced from hardwood includes a number of gelatinous layers containing a large amount of cellulose microfibrils, and serves to maintain straight trunks by applying tensile stress to them. On the other hand, compression wood produced from softwood growing on slopes, as used for samples in the present study, is generated on the side of the trees facing the valley. Compression or tension wood produced from softwood species includes a number of short tracheids containing a large amount of lignin, and applies compressive stress to maintain a straight trunk. Cellulose microfibrils are rigid, and lignin is more viscous than cellulose. Therefore, the following assumption is reasonable. Tension wood of hardwood pulls up the tree body by increasing the amount of cellulose having the rigid property, and compression wood of softwood pushes up the tree body by increasing the amount of lignin having viscosity properties of more than cellulose. Furthermore, the roots of trees are a region subjected to large stress, and studies on their characteristics have been conducted in recent years. The characteristics of roots are very interesting because they are the only region that contacts the earth and supports the whole tree. Previous studies on softwood suggest that the microfibril angle of the roots in the early developmental stage is smaller and Young's modulus is higher as compared to the trunks [17, 18]. This is presumably because the roots of a tree are required to grow at a faster rate than the trunk to provide the tree with support, as it has already grown large by this stage, and maintain its stability. As this example shows, trees change their chemical components and tissue structures to optimize support and maintain balance in their growing environments.

On the other hand, a large number of leaves grow on branches against gravity to perform photosynthesis. The branches bow and bounce back when the rain is falling or wind is blowing so that the surfaces of the leaves face the sun. Therefore, the bases of branches may be subjected to greater stress than trunks and other parts of the branches in terms of structural dynamics. However, lignin contained in trees, softwood ones as used in the present study in particular, is transitioned to glass at approximately room temperature, although it is more viscous than cellulose microfibrils, as shown by previous studies [19]. In general, the polymer materials in vitreous state are known to be more brittle and tolerant to breaking strain than materials in rubber or glass-transition states [15]. It is difficult for the trunks to support the branches, while resisting gravity or in the rain and winds. The trees may accumulate specific constituents in the bases of their branches, expected to be subjected to the greatest stress in terms of structural dynamics, to promote the strength of part of vitrified lignin and increase its elasticity and viscosity by activating it so that the branches can tolerate the stress. It is suggested that E', E'', and tan δ in the bases of the branches at approximately room temperature, the latter two in particular, increased, and dynamic relaxation as a result of the glass transition of part of lignin was noted at room temperature.

To gain further knowledge in relation to clarification of the support mechanism and physical properties of the branches, further studies such as carrying out studies for other tree species will be needed.

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