J Wood Sci (2011) 57:1–6
DOI 10.1007/s10086-010-1133-2

ORIGINAL ARTICLE

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New piezoelectric moduli of wood: d_{31} and d_{32}

Received: January 21, 2010 / Accepted: May 17, 2010 / Published online: August 18, 2010

Abstract This article reports the piezoelectric moduli of wood d_{31}, d_{32}, and d_{36}. The piezoelectric moduli of wood d_{31} and d_{32} have not been previously reported, although there has been much research on the d_{14} and d_{25} moduli of wood. The moduli d_{31}, d_{32}, and d_{36} were measured carefully because their absolute values were considerably smaller than those of d_{14} and d_{25}. For Softwoods, d_{36} values were mostly negative, whereas the values for hardwoods had either positive or negative values. The other moduli, d_{31} and d_{32}, were a mixture of positive and negative values in softwoods and hardwoods. The existence of d_{31} and d_{32} suggests the presence of an electrical polarity of the cellulose crystal in the fiber direction of the wood. The polarities of d_{31} and d_{32} became clear from wood in the outer part of the trunk, where the crystallinity of cellulose is large and the alignment of the crystals becomes parallel to the fiber direction.

Key words Wood · Piezoelectric modulus · d_{31}, d_{32}, and d_{36} · Cellulose-I crystal · Piezoelectric polarity

Introduction

The piezoelectric effect of wood is caused by the crystalline structure of native cellulose fibrils. The cellulose micelles in wood are oriented mostly along the fiber axis, and so wood belongs to symmetry class D_∞.[1] Accordingly, wood has only two piezoelectric moduli, d_{14} and d_{25}, whose magnitudes are usually the same but with opposite polarities; they have been measured as the piezoelectric moduli of wood.[2–4] The absolute values, however, are not equal for d_{14} and d_{25} in some cases. Considering the structural anisotropy of wood in the radial and tangential directions, wood would seem to have the piezoelectric modulus d_{36} also.[5] The magnitude of d_{14} and d_{25} of wood is the order of 10^{-13} C/N and is about 1/20 of the piezoelectric modulus d_{11} of quartz. The absolute value of d_{36} obtained in this experiment was almost a half of those for d_{14} and d_{25}. This article reports the piezoelectric moduli of wood d_{31}, d_{32}, and d_{36} in relation to tree growth. The existence of the new moduli d_{31} and d_{32} of wood was confirmed.

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Part of this work was presented at the 14th Annual Meeting of the Cellulose Society of Japan, Shizuoka, July 2007, and in Cellulose Communications 2003;10:119–123

Materials and methods

Definition of piezoelectric moduli of wood d_{31}, d_{32}, and d_{36}

The cellulose-I crystal belongs to the monoclinic system with symmetry group C_2,[6] which has eight piezoelectric moduli: d_{14}, d_{15}, d_{24}, d_{25}, d_{31}, d_{32}, d_{33}, and d_{36}.[7] Piezoelectric moduli of wood are determined by the symmetry of the wood structure, as shown in Fig. 1. In rectangular coordinates, z-, x-, and y-axes coincide with the longitudinal, radial, and tangential directions of the tree trunk, respectively. Figure 1b shows the specimens for the measurements of $d'_{31}(0)$, $d'_{31}(45)$, and $d'_{31}(90)$.

The transformed piezoelectric modulus d'_{31} for rotation through an angle θ about the z-axis is given by:[8]

$$d'_{31} = d'_{31}(\theta) = \cos^2\theta d_{31} + \sin^2\theta d_{32} + \sin\theta\cos\theta d_{36} \quad (1)$$

The modulus d_{36} is the coefficient of the polarization in the z-axis subjected to shear stress in the xy plane. The modulus d_{31} is that in the z-axis by uniaxial stress in the x direction and the modulus d_{32} is that in the z-axis by uniaxial stress in the y direction.

The measured value of $d'_{31}(\theta)$ is made up of the contributions of d_{31}, d_{32}, and d_{36} as shown in Eq. 1, and it depends on the rotation angle about the z-axis. The characteristic moduli of piezoelectricity before the transformation about the z-axis rotation, d_{31}, d_{32}, and d_{36}, were determined from Eq. 1

by using the experimental values of $d'_{31}(0)$, $d'_{31}(22.5)$, $d'_{31}(45)$, $d'_{31}(67.5)$, and $d'_{31}(90)$, which are the d'_{31} values in the directions 0°, 22.5°, 45°, 67.5°, and 90° from their radial direction in the cross section. Optimization of the characteristic values of d_{31}, d_{32}, and d_{36} was done using the least-squares method. Data sets of five specimens at the angles of 0°, 22.5°, 45°, 67.5°, and 90° from the radial direction were used in the calculation. Finally, the normal equations were solved by minimizing the residual sum of squares of the measured and predicted values.

Specimens

Seven kinds of softwoods and eight kinds of hardwoods were used, as shown in Table 1. The top and bottom directions of the trunks were confirmed when the tree was felled. Specimens of Yezo spruce, Douglas fir, Japanese cedar, and *Prunus subhirtella* were prepared from pieces of large lumber 1–2 m in length in the fiber direction, including the pith and bark.

Specimens for the measurements of $d'_{31}(0)$, $d'_{31}(22.5)$, $d'_{31}(45)$, $d'_{31}(67.5)$, and $d'_{31}(90)$ were cut at angles 0°, 22.5°, 45°, 67.5°, and 90° from the radial direction in the cross section. The typical size of a specimen was 2.5 cm (length) × 0.8 cm (width) × 0.1 cm (thickness). Square aluminum foil was bonded on the specimen surfaces by silicone paste. Aluminum ribbons were attached to the electrodes as lead wires. The annual ring number of the samples noted here was defined from the center. Multiple annual rings were included in the electrode plane because the electrode size was larger than the annual ring breadth. The number of annual rings included in the electrode area varied from about 5 to 15. Specimens for measuring d_{25} were cut at an angle of 45° from the fiber direction in the radial plane, following the same procedure as that reported before.[5]

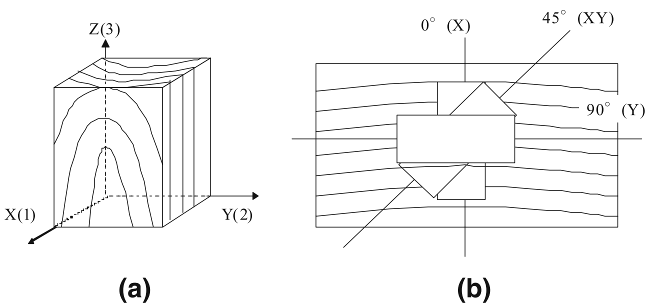

Fig. 1. Definition of piezoelectric moduli. **a** Rectangular coordinates assigned to wood, **b** the cutting of wood specimens for d'_{31}

Measurements of piezoelectric modulus

Specimens were dried in a vacuum chamber for 2 h and were kept in the absolutely dry condition with phosphorus pentoxide for several days. The piezoelectric moduli were measured at four frequencies, i.e., 13, 26, 52, and 104 Hz, using a Rheolographsolid (Toyo Seiki, Tokyo, Japan) at a temperature of 20ºC. The piezoelectric modulus of a specimen was determined as the mean value for the four frequencies because the modulus was almost constant in the frequency range used, as shown in Fig. 2. The piezoelectric measurements were done twice by changing the face and back planes of the specimen; this was done because by changing the face and back of the electrodes, the direction of the piezoelectric polarization could be confirmed more certainly. Some specimens showed the same polarization, although the face and the back were changed. We consider that the data are within the error of the device used, but the values were very small, i.e., of the order of 10^{-16} C/N. Accordingly, we plotted the data as zero in Figs. 3–8.

The following three kinds of measurements were conducted:

Table 1. Specimens and values of piezoelectric moduli of wood d_{31}, d_{32}, d_{36}, and d_{25}

Species	Position of specimen in the trunk			Stand of tree	d_{31}	d_{32}	d_{36}	d_{25}
	Height (m)	Direction	Ring number		($\times 10^{-15}$C/N)			
Softwood								
Chamaecyparis obtusa (Japanese cypress)	2	North	50~65	(1)	–1.45	–0.63	–25.82	102
Sequoia sempervirens (redwood)	3.5	North	35~50	(1)				39.4
Pinus taeda (loblolly pine)	–	ldf	25~30	(1)	–1.39	1.51	–2.20	88.2
Pinus palustris (long leaf pine)	4	North	50~60	(1)	2.03	–0.90	–11.70	54.2
Picea jezoensis (Yezo spruce)	–	ldf		(4)				
Pseudotsuga menziesii (Douglas fir)	–	ldf		(4)				
Cryptomeria japonica (Japanese cedar)	–	ldf		(4)				
Hardwood								
Kalopanax pictus	1	North	11–37	(2)				75.8
Quercus acuta	1	North	9–28	(2)	–3.55	–0.53	2.96	13.5
Prunus jamasakura	1	North	10–41	(2)				99.6
Carpinus tschonoskii	1	North	35–45	(2)	2.00	–3.21	4.72	118
Betula grossa	1	North	32–57	(3)	–3.10	2.82	8.80	114
Acer mono	1	North	19–37	(2)	1.18	–2.48	–6.99	51
Quercus myrsinaefolia	1	North	20–30	(2)	–2.10	1.19	–4.51	102
Prunus subhirtella	–		25–42	(4)	1.90	–0.64	1.09	

Tree stands were: (1), Tama Forest Garden (Governmental FFPRI of Japan); (2), Kamiatago Shizuoka University experimental forest; (3), Nakakawane Shizuoka University experimental forest; (4), unknown ldf, least diameter direction

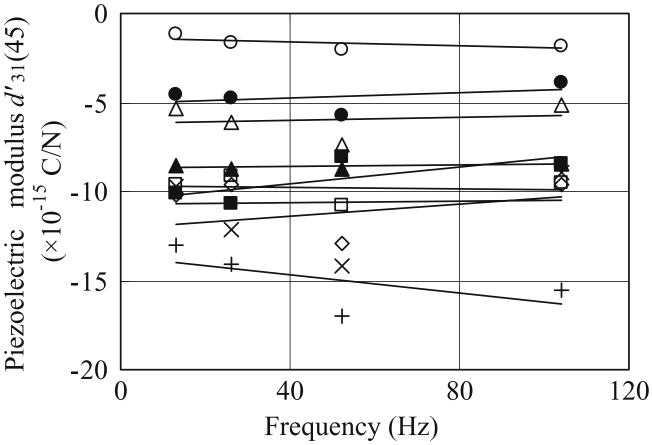

Fig. 2. Frequency response of piezoelectric modulus $d'_{31}(45)$. Specimens are from the south-facing part of a Japanese cypress tree. Regression lines for nine different specimens are shown. Identical symbols indicate experimental values of a specimen

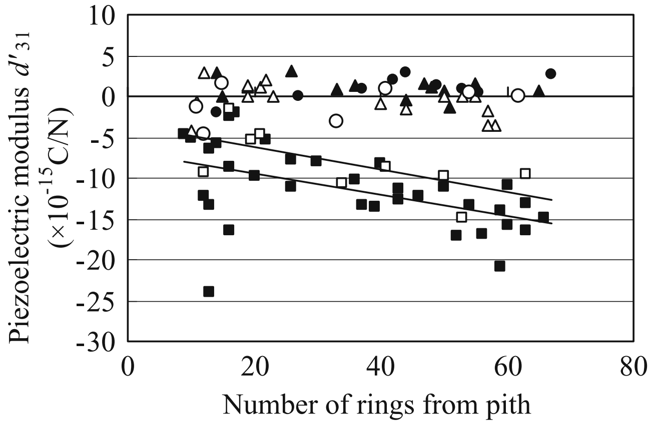

Fig. 3. Variation of piezoelectric modulus d'_{31} of Japanese cypress from pith to bark. *Solid and open symbols* are from north- and south-facing samples, respectively. *Circles*, $d'_{31}(0)$; *triangles*, $d'_{31}(90)$; *squares*, $d'_{31}(45)$; *lines*, regression lines of $d'_{31}(45)$

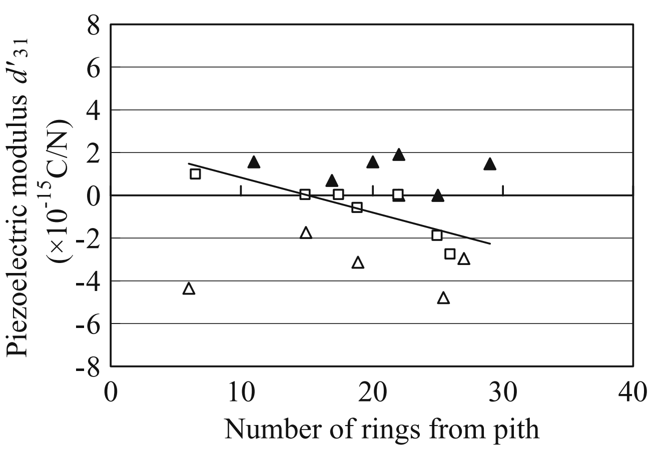

Fig. 4. Variation of piezoelectric modulus d'_{31} of *Quercus myrsinaefolia* from pith to bark. *Open triangles*, $d'_{31}(0)$; *solid triangles*, $d'_{31}(90)$; *open squares*, $d'_{31}(45)$; *solid line*, regression line of $d'_{31}(45)$

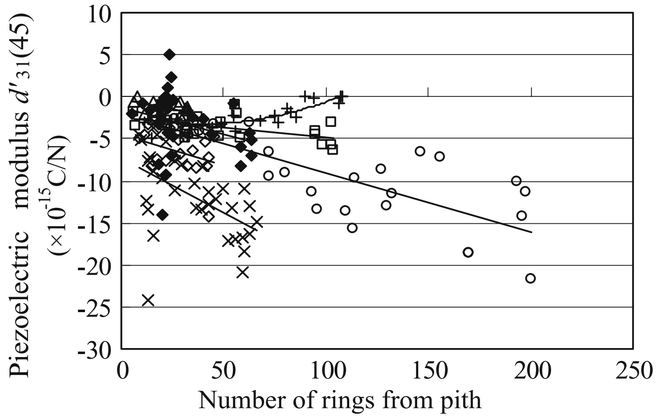

Fig. 5. Variation of piezoelectric modulus $d'_{31}(45)$ of softwoods from pith to bark. *Open triangles*, loblolly pine; *open diamonds*, redwood; *crosses*, Japanese cypress; *solid diamonds*, long leaf pine; *open circles*, Yezo spruce; *open squares*, Japanese cedar; *plus signs*, Douglas fir; *solid lines*, regression lines

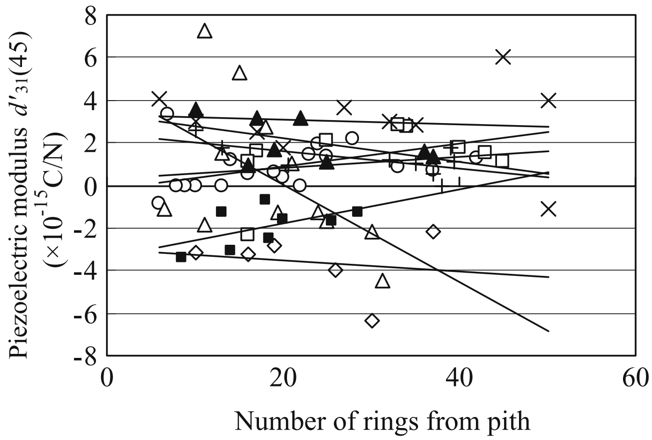

Fig. 6. Variation of piezoelectric modulus $d'_{31}(45)$ of hardwoods from pith to bark. *Open circles*, *Prunus subhirtella*; *open squares*, *Carpinus tschonoskii*; *solid triangles*, *Kalopanax pictus*; *crosses*, *Betula grossa*; *open triangles*, *Quercus acuta*; *open diamonds*, *Acer mono*; *plus signs*, *Prunus jamasakura*; *solid squares*, *Quercus myrsinaefolia*; *solid lines*, regression lines

1. The piezoelectric moduli of wood $d'_{31}(0)$, $d'_{31}(45)$, and $d'_{31}(90)$ were measured from the pith to the bark.
2. The distribution of piezoelectric polarization was measured by rotating the z-axis in the xy plane by 0°, 22.5°, 45°, 67.5°, and 90° using the outside specimens whose annual ring number is large.
3. Additional measurements of d_{25} were conducted to obtain reference data.

Results and discussion

Piezoelectric modulus of wood $d'_{31}(\theta)$

Figure 3 shows the variations of d'_{31} of Japanese cypress, $d'_{31}(45)$, $d'_{31}(0)$, and $d'_{31}(90)$, in relation to the annual ring number from the pith. The moduli $d'_{31}(45)$ showed similar

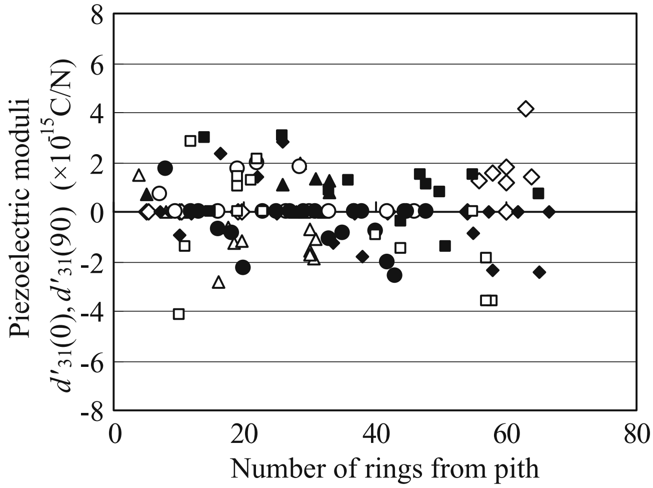

Fig. 7. Variations of piezoelectric moduli $d'_{31}(0)$ and $d'_{31}(90)$ of softwoods from pith to bark. *Open* and *solid symbols* are data for $d'_{31}(0)$ and $d'_{31}(90)$, respectively. *Triangles*, loblolly pine; *diamonds*, long leaf pine; *circles*, redwood; *squares*, Japanese cypress

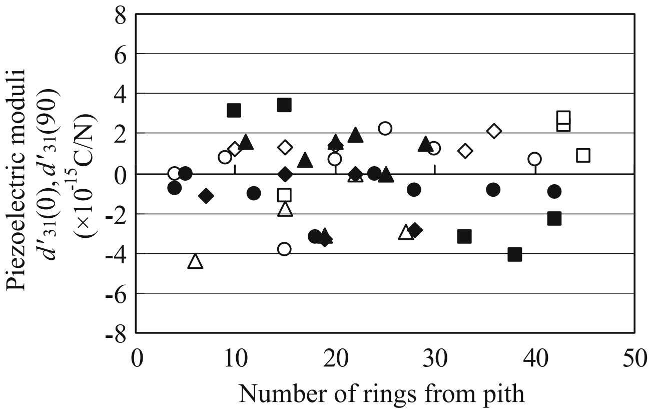

Fig. 8. Variations of piezoelectric moduli, $d'_{31}(0)$ and $d'_{31}(90)$ of hardwoods from pith to bark. *Open* and *solid symbols* are data for $d'_{31}(0)$ and $d'_{31}(90)$, respectively. *Diamonds*, *Acer mono*; *squares*, *Carpinus tschonoskii*; *circles*, *Prunus subhirtella*; *triangles*, *Quercus myrsinaefolia*

changes for both south- and north-facing parts of the disk, and all such values were negative. The absolute value of $d'_{31}(45)$ was of the order of 10^{-14} C/N and tended to be larger in the outer part of the trunk. Those of $d'_{31}(0)$ and $d'_{31}(90)$ were very small compared with $d'_{31}(45)$. Some values were positive and some negative for both $d'_{31}(0)$ and $d'_{31}(90)$. The sign of $d'_{31}(0)$ was mostly positive and that of $d'_{31}(90)$ was mostly negative in the outer part of the trunk where the annual ring number is large.

Figure 4 shows the variations of d'_{31} of *Quercus myrsinaefolia*,an example of a hardwood. The trend is different from that of Japanese cypress, which is a softwood. Although the absolute value of $d'_{31}(45)$ of Japanese cypress was larger than those of $d'_{31}(0)$ and $d'_{31}(90)$, this trend was not shown in the case of *Quercus myrsinaefolia*. The sign of $d'_{31}(0)$ was negative and that of $d'_{31}(90)$ was mostly positive.

Figure 5 shows the variations of $d'_{31}(45)$ for the seven species of softwoods shown in Table 1. The absolute value of $d'_{31}(45)$ tended to increase with the increase of annual ring number in each softwood. The sign of $d'_{31}(45)$ was negative, except for the long leaf pine at annual ring numbers around 20. The specimens of the long leaf pine shown in Fig. 5 were prepared from the north-facing part at 4 m high. Additional measurements were conducted to confirm the above exceptional data for the same trunk of long leaf pine; this time, the specimens were prepared from the south-facing part at 4 m high and the north-facing part at 10-m high. These additional measurements yielded negative values for $d'_{31}(45)$, similar to the other softwoods.

The above results show that the signs of most $d'_{31}(45)$ values were negative, except for a few cases in the long leaf pine. The cause of the exceptional cases is not clear, but some growth change under unusual environmental conditions could have affected the values. Accordingly, our results suggest that the signs of $d'_{31}(45)$ values for softwoods are negative.

Figure 6 shows the variations of $d'_{31}(45)$ for the eight hardwood species listed in Table 1. They were prepared from north-facing parts of the trunk at 1 m high from the ground, except for *Prunus subhirtella*. There were large differences in the modulus distributions between the softwoods and the hardwoods. In the hardwoods, the signs of some $d'_{31}(45)$ values were negative and some positive. The signs were positive for *Prunus jamasakura* and *Kalopanax pictus*, and mostly positive for *Carpinus tschonoskii, Betula grossa*, and *Prunus subhirtella*. For *Acer mono* and *Quercus myrsinaefolia*, the $d'_{31}(45)$ values were all negative. The values for *Quercus acuta* were sometimes positive and sometimes negative.

Figures 7 and 8 show the variations of $d'_{31}(0)$ and $d'_{31}(90)$ for softwoods and hardwoods, respectively. The signs of the moduli were a mixture of negative and positive, and the trend depended on the tree species. For softwoods, the absolute values of $d'_{31}(0)$ and $d'_{31}(90)$ were considerably smaller than those of $d'_{31}(45)$. The signs of the $d'_{31}(0)$ and $d'_{31}(90)$ values were different for each tree species. In the loblolly pine and the Japanese cypress, the signs of $d'_{31}(0)$ were mostly negative and those of $d'_{31}(90)$ were mostly positive. In the long leaf pine and the redwood, the signs of $d'_{31}(0)$ were mostly positive and those of $d'_{31}(90)$ were mostly negative. For the hardwoods, the signs of $d'_{31}(0)$ of *Acer mono*, *Carpinus tschonoskii*, and *Prunus subhirtella* were mostly positive and those of $d'_{31}(90)$ were mostly negative. For *Quercus myrsinaefolia*, the sign of $d'_{31}(0)$ was negative and that of $d'_{31}(90)$ was positive.

Characteristic moduli of piezoelectricity of wood d_{31}, d_{32}, and d_{36}

The piezoelectric moduli of wood d_{31}, d_{32}, and d_{36} were calculated using the least-squares method on Eq. 1. The values of $d'_{31}(\theta)$ were obtained from the experiments using specimens from the outside parts of the trunks, as shown in Table 1, because the crystallinity and the piezo-

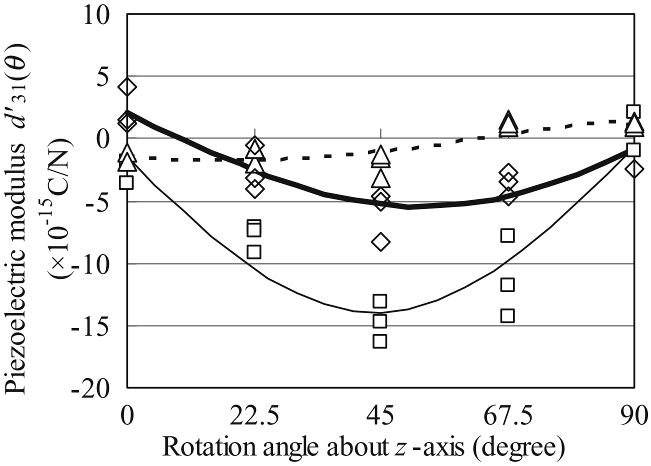

Fig. 9. Anisotropy in piezoelectric modulus $d'_{31}(\theta)$ and approximate curves of $d'_{31}(\theta)$ for softwoods. *Symbols* are experimental values. *Lines* indicate approximated curves. *Squares*, Japanese cypress; *diamonds*, long leaf pine; *triangles*, loblolly pine; *fine line*, Japanese cypress; *broad line*, long leaf pine; *dotted line,* loblolly pine

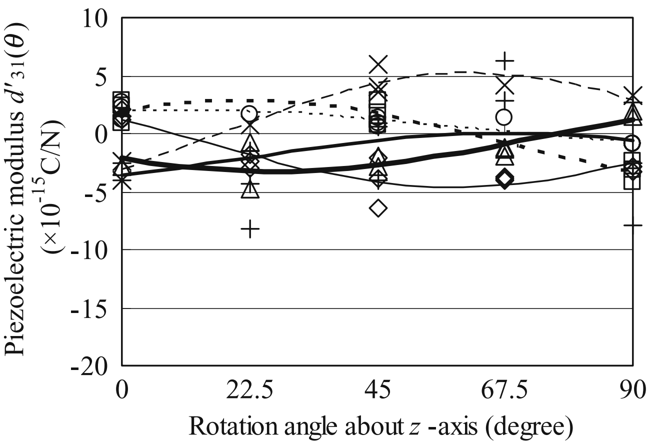

Fig. 10. Anisotropy in piezoelectric modulus $d'_{31}(\theta)$ and approximate curves of $d'_{31}(\theta)$ for hardwoods. *Symbols* are experimental values. *Lines* indicate approximated curves. *Diamonds*, *Acer mono*; *triangles*, *Quercus myrsinaefolia*; *circles*, *Prunus subhirtella*; *squares*, *Carpinus tschonoskii*; *crosses*, *Betula grossa*; *plus, Quercus acuta*; *fine line*, *Acer mono*; *superbroad line*, *Quercus myrsinaefolia*; *fine dotted line*, *Prunus subhirtella*; *broad dotted-line*, *Carpinus tschonoskii*; *broken line*, *Betula grossa*; *broad line, Quercus acuta*

electric modulus of wood become larger with growth.[9] The details of the specimen positions in the trunk are shown in Table 1.

Data for which the polarization did not change by turning the face and back of the specimens, especially in the directions 0° and 90° from the radial direction, are not suitable for inclusion in the quantitative evaluation of the piezoelectric moduli d_{31}, d_{32}, and d_{36}. Accordingly, such data were excluded from the calculations.

Figures 9 and 10 show the piezoelectric moduli $d'_{31}(\theta)$ for softwoods and hardwoods, respectively, plotted against the angle θ. For softwoods, the signs of the moduli were distributed in the negative region for most angles, and the absolute values were maximum at $\theta = 45°$. The signs of $d'_{31}(45)$ were negative in these cases. For hardwoods, the angular distributions were considerably different among the tree species. The signs of $d'_{31}(\theta)$ were scattered both on the positive and negative sides.

The results of the three characteristic moduli estimated from the inverse calculation are shown in Table 1. The values of d_{25} for the eleven species measured using specimens with relatively large annual ring numbers are also shown in Table 1. The absolute values of d_{31}, d_{32}, and d_{36} were considerably smaller than those of d_{25}.

The piezoelectric phenomenon of wood depends on the structure of cellulose crystals and the orientation of cellulose crystals in the wood. Generally, wood is classified in symmetry class D_∞ because it is assumed that the cellulose micelles in wood are oriented parallel to the z-axis, that the distribution of each micelle is random in the positive and negative directions of the z-axis, and that the distribution of micelles in the xy plane is quite random. In this case, the number of piezoelectric moduli for wood becomes only two, i.e., d_{14} and $d_{25,}$ from the requirements of the symmetry class.

Although much research has been carried out on d_{14} and d_{25} of wood, the existence of moduli d_{31}, d_{32}, and d_{36} has not been reported. However, if we hypothesize that there is a polarity in the cellulose-I crystal in the axial direction and that the orientation of the cellulose micelles in the wood cell walls has an imperfect symmetry, then piezoelectric moduli d_{31}, d_{32}, and d_{36} would be observed.

In our experiments, the magnitudes of the piezoelectric moduli varied for softwoods and hardwoods. For softwoods, the polarities of d_{36} were negative and the absolute values were always larger than those of d_{31} and $d_{32.}$ However, for hardwoods, some values of d_{36} were positive and some negative, and there were two cases in which the absolute values of d_{31} were larger than those of d_{36} for hardwoods, i.e., for *Quercus acuta* and *Prunus subhirtella*, as shown in Table 1.

First, we confirmed the existence of d_{36}. Piezoelectric moduli for such an assembled system of crystallites can be calculated by taking an average of the moduli of the individual crystallites.[2,10] Modulus d_{36} does not appear if the orientation of the cellulose crystal is isotropic around the longitudinal axis of the wood; however, mechanical and physical properties of wood are generally not isotropic in the cross section. Anisotropies in the elastic constants in the cross section are the cause of piezoelectric constant d_{36} because the piezoelectric effect is a combination of the elastic property and polarization due to dislocation of electric charge. A breaking of the symmetry would be the reason for the existence of d_{36}. The existence of d_{36} would depend on the differences in the piezoelectric effects between the radial and tangential directions. Softwoods showed mostly negative polarization, whereas for the hardwoods, there were both negative and positive cases in the polarizations of d_{36}.

Next, the existence of d_{31} and d_{32} was confirmed as the piezoelectric moduli of wood in this experiment. Figure 11 shows the relation between the alignment of cellulose molecules in a cellulose-I crystal and the polarity.

The orientation of each cellulose molecule in the cellulose-I crystal has traditionally been considered to be anti-

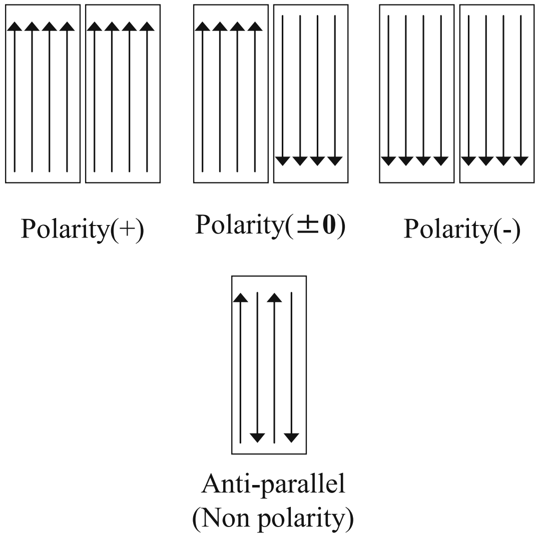

Fig. 11. The relation between alignment of cellulose molecules in a cellulose-I crystal and electric polarity. *Arrows* show the directions of cellulose molecules in a cellulose-I crystal

parallel, based on the X-ray model of Mayer-Misch.[11] Such moduli as d_{31} and d_{32} must disappear by the requirement of structural symmetry. In order to explain the existence of moduli d_{31} and d_{32}, at the very least, polarity must exist for individual cellulose micelles, i.e., the direction of the cellulose molecules in each micelle must be parallel.

In our experiments, the polarities of some values for piezoelectric moduli d_{31} and d_{32} were positive and some negative. This phenomenon can be explained by assuming an imbalance in the directions of the individual micelles in the wood cell walls. If the number of micelles directed in the upward and downward directions is equal, such piezoelectric moduli must disappear. The polarity of the piezoelectric modulus depends on the imbalance of the distribution of the micelle directions.

Resent research on cellulosic substances using NMR measurements has supported the parallel orientation of the molecular chains.[12,13] If the direction of the cellulose molecules in the cellulose-I crystals of wood is also parallel, this provides a mechanism explaining why the piezoelectric moduli of wood d_{31} and d_{32} have polarity. The existence of the new piezoelectric constants d_{31} and d_{32} of wood also suggests the existence of a pyroelectric effect in the fiber direction of wood.

Conclusion

The piezoelectric moduli of wood d_{31}, d_{32}, and d_{36} were observed.

The following conclusions were obtained:

1. The existence of the piezoelectric modulus d_{36} was confirmed in some softwoods and hardwoods; the magnitudes of d_{36} were considerably smaller than those of d_{14} and d_{25}. The polarity of d_{36} was mostly negative for softwoods and either positive or negative for hardwoods.
2. The piezoelectric moduli d_{31} and d_{32} were observed; however, their magnitudes were very small (of the order of 10^{-15} C/N) and had positive or negative polarization.
3. The existence of the piezoelectric moduli d_{31} and d_{32} supports the suggestion that the molecule orientations in the cellulose-I crystals of wood are parallel.

Acknowledgments The authors are sincerely grateful to Mr. Yushirou Miwa, Tama Forest Science Garden of Forestry and Forest Products Research Institute, for kindly providing the disks of softwoods; to Mr. Taichi Kajikawa and Mr. Hayahito Yazawa, Shizuoka University Forest, for felling trees; and to Mr. Shunji Hayamura, Shizuoka University, for photographing the trees. The authors also thank Dr. Tomoyuki Fujii, Forestry and Forest Products Research Institute; Dr. Masahisa Wada, the University of Tokyo; and Dr. Shinji Hirai, emeritus professor of the University of Tokyo, for their helpful suggestions and comments.

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