Studies on Dielectric Behavior of Bound Water in Timber in the High Frequency Region

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Introduction

It has been well-known up to this time that the dielectric properties of timber remarkably depend upon the water contained. Recently, a new industry of utilizing the high frequency electric waves as a heat source for drying timbers or for gluing wood pieces together was developed in this country,(1) and many factors involved in the process were examined, *e. g.*, the amount of water contained, the temperature of the treatment and the wave-length of electric wave to be employed. Indeed, it is often regarded important as well as interesting to investigate why the water contained in timber has such a predominant effect upon its dielectric properties.

In the present paper, the author has intended to report the dielectric behavior of timber in the high frequency region and its possible interpretation of the results. Special attentions were paid to the measurements of the microwave region, where remarkable dielectric dispersion and absorption are expected to occur. And in comparing the dielectric behavior of pure water and that of bound water in timber, we hoped to obtain some useful informations about the state of the latter.

Experimental Method

The measurements of complex dielectric constants were carried out

1) at 3×10^3 M. C. by the free wave method(2,3)
2) *ditto* by the wave guide method(4,5)
3) at 300 M. C. by the impedance bridge method(6)
4) in the region of 0.2—15 M. C. by the dynatron method(7)
5) d. c. conductivity by the usual galvanometer method.

The timbers of the species *tilia* (product in Hokkaido) were chosen as the experimental materials. The wood pieces to be tested were prepared in three different ways, *i. e.*, (a) cut perpendicular to the wood vessels, (b) to the medullary rays and (c) parallel to the both. The samples which were dried for several hours in a drought at the temperature of 100° were assumed as the standard material for zero-water-content. They were dried in a bath and then put into a vessel, the humidity of which was kept constant by the use of H_2SO_4 solution. The amount of absorbed water was determined with a chemical balance.

Experimental Results

The measurements of dielectric constant and loss factor were made in the region of 0.2-3000 M. C. for the samples of various water-contents (from 0 to 14 % at 300 and 3000 M. C.; from 0 to 20 % in the region of 0.2—15 M. C.). The results were shown in Figs. 1-6, where ε' denotes dielectric constant, ε'' loss factor, σ

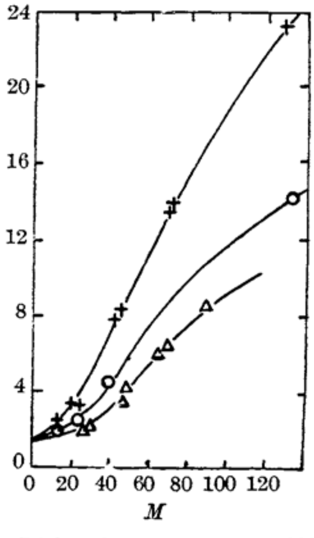

Fig. 1.—Dielectric constant at 3000 M. C. (room temp.): ×, the electric field ‖ wood vessels; ○, the electric field ‖ medullary rays; △, the electric field ⊥ wood vessels and ⊥ rays.

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(1) "Theory and Application of High Frequency Phenomena," ed. by Asami (1950), Rasearch Institute of Applied Electricity, Hokkaido University.

(2) Yasumi, This Bulletin, **24**, 53 (1951).

(3) This measurement was made with the help of Mr. K. Nukazawa, Research Institute of Applied Electricity, Hokkaido University.

(4) Matsumoto and Suzuki, *Bull. Technology* (Hokkaido University) **1**, 138 (1949).

(5) This measurement was carried out in the collaboration with Prof. Suzuki of the Department of Electric Engineering, Faculty of Technology, *ditto*.

(6) Morita, *J. Inst. Elec. Eng.* (Japan)., **65**, 59 (1946).

(7) Nishi and Sakamoto, *ibid.*, **59**, 699 (1940).

conductivity, f frequency, ω angular frequency and M water-content (in weight percentage), respectively.

In Figs. 1 and 2, dielectric constants and loss-factors measured at 3000 M. C. are plotted against the moisture-content, while in Fig. 3 the results obtained at 300 M. C. are shown. In all these diagrams the variations in ε' and ε'' due to the inhomogeneity of the material are clearly observed.

In Figs. 4 and 5 are shown the results obtained for a wide range of frequencies——in Fig. 5 the direct current measurement is also included. In Fig. 4 the increase in ε' with the increase of water-content is found to decrease with the increase in frequency. It is further to be noted that these increases in value of ε' are not marked in the range of 0 to 5 %. Moreover, it is interesting that the curves

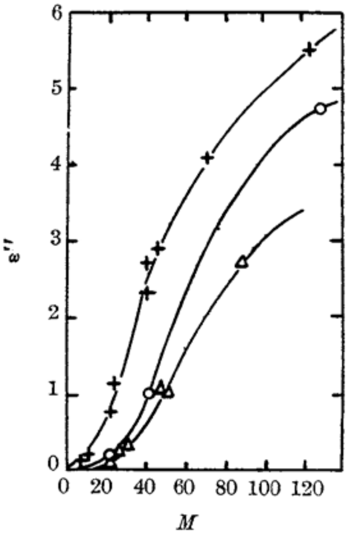

Fig. 2.—Loss factor at 3000 M. C. (room temp.) ×, ○, △, *ditto.*

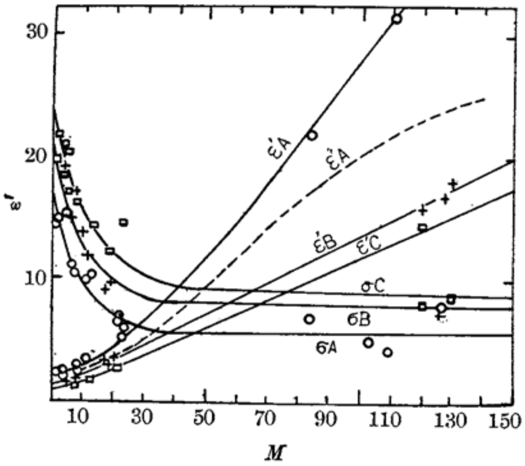

Fig. 3.—Dielectric constant and conductivity at 300 M. C. (full line) and at 3000 M.C. (broken line) (temp. 15°±2°C.): A, the electric field $\parallel$ the wood vessels; B, the electric field $\parallel$ the medullary rays; C, the electric field $\perp$ vessels and rays.

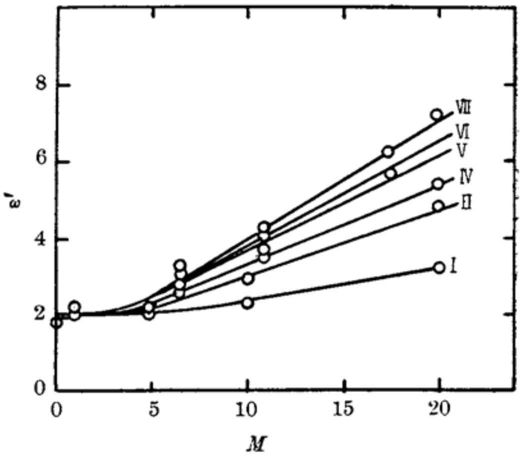

Fig. 4.—Dielectric constant and water-content (15°C.): I, 3×10^9 cycles; II, 3×10^8 cycles; IV, 1.0×10^7 cycles; V, 3×10^6 cycles; VI, 1×10^6 cycles; VII, 0.4×10^6 cycles.

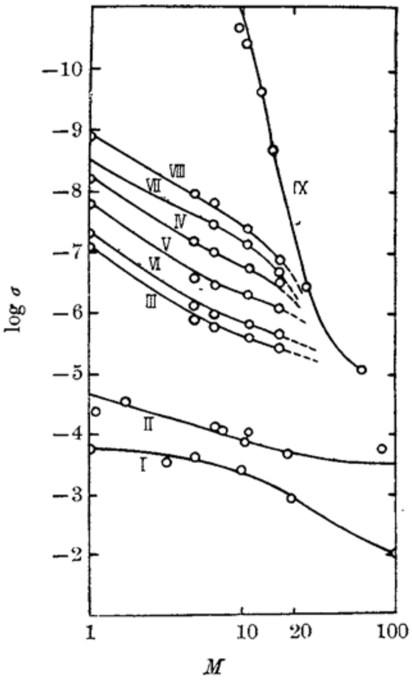

Fig. 5. — Conductivity and water-content (15°C.): I, 3×10^9 cycles; II, 3×10^8 cycles; III, 15×10^6 cycles; IV, 10×10^6 cycles; V, 3×10^6 cycles; VI, 1×10^6 cycles; VII, 4×10^5 cycles; VIII, 2×10^5 cycles; IX, d. c.

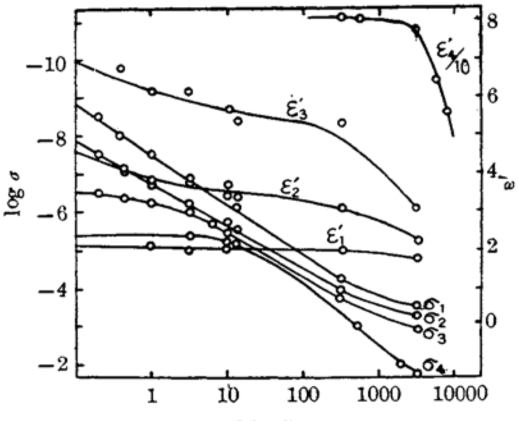

Fig. 6.—Dielectric constant and conductivity (15°C.).

ε'_1, σ_1	water-content	3.5%
ε'_2, σ_2	〃	10%
ε'_3, σ_3	〃	20%
ε'_4, σ_4	pure water	

of log σ *versus* M in Fig. 5 are similar to those of the dried carrot obtained by Dunlap and Makover⁽⁸⁾.

The values of $\mathcal{E}'$ and $\log\sigma$ of the samples of the same water-content are plotted against the frequency in Fig. 6. The dielectric constants of the samples of the 10 % and 20 % water-contents decrease with the increase in frequency, while log σ generally increases with the increase of frequency. On closer examination, it appears that the inflection points of the curves for low-water-content shift to the side of lower frequency than those for high-water-content and that for pure water.

Discussion

To begin with the discussion, we must admit that our material of study, timber, is a very complicated substance to which no heterogeneous theory of dielectrics is quite applicable. Therefore we will not venture to give any complete explanation for the results observed but rather wish to present some useful notes connected with this topic. First of all, we will consider the dielectric behavior of pure water. (See the curves of $\mathcal{E}'_4$ and σ_4 in Fig. 6.) The relation between dielectric constant (or loss factor) and the frequency was treated theoretically by Debye⁽⁹⁾ on the basis of dipole rotation. If we assume the Onsager's inner field instead of the Clausius-Mosotti's relation which was adopted in the Debye's original theory, the dielectric quantities will be expressed as follows:

$$\left.\begin{aligned} \mathcal{E}'-i\mathcal{E}'' &= \mathcal{E}_{(\infty)}+\frac{\mathcal{E}_{(0)}-\mathcal{E}_{(\infty)}}{1+i\omega\tau} \\ \text{and}\qquad \sigma &= \frac{\omega\mathcal{E}''}{4\pi} \end{aligned}\right\} \tag{1}$$

where $\mathcal{E}_{(0)}$ and $\mathcal{E}_{(\infty)}$ are dielectric constants at $\omega=0$, and $\omega=\infty$ respectively, τ is the relaxation time which differs from that of Debye by a constant factor.

We put the values $\mathcal{E}_{(0)}=81$, $\mathcal{E}_{(\infty)}=1.8$ and $\tau=9.4\times10^{-12}\,\text{sec}^{-1}$ (at 15°) into Eq. (1), and the calculated results are found to be in good accord with the experimental data for angular frequencies ω above 50 M. C.

As often pointed out, the following equation holds in the low frequency range.

$$\log\sigma = 2\log f+\text{const.} \tag{2}$$

If the relation that $\omega<\dfrac{1}{\tau}$ is assumed, then Eq. (2) can easily be derived from Eq. (1). If water has an appreciable d. c. conductivity, σ_{ion} which arises from the movement of ions, σ is expressed as follows:

$$\sigma = \frac{(\mathcal{E}_{(0)}-\mathcal{E}_{(\infty)})}{4\pi}\cdot\frac{\omega^2\tau}{1+\omega^2\tau^2}+\sigma_{\text{ion}} \tag{3}$$

(σ and σ_{ion} in e. s. u.)

The experimental values of σ in the region of frequencies less than 50 M. C. are found to agree with that of Eq. (3).

The dielectric constant of timber in general increases with the increase in water-content. Further, it is to be noted that the dispersion region of the dielectric constant of timber (10 %—20 % water-content) shifts to the side of the longer wave length than that of pure water⁽¹⁰⁾. This fact is an indication that the relaxation time of bound water in timber has a larger value than that of pure water. According to Debye, the relaxation time τ is related to internal viscosity η by the following equation.

$$\tau = \frac{4\pi\eta a^3}{kT} \tag{4}$$

where a is the radius of rotating molecule. If the internal viscosity η of bound water happens

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(8) Dunlap and Makover, *J. Phys. Chem.*, **49**, 601 (1945).

(9) Debye, "Polare Molekeln," Leipzig, 1929, s. 108.

(10) The dielectric behavior of ethyl alcohol absorbed by silica gel was studied by Higuti and it was found that dielectric dispersion shifted to a longer wave length than that of pure ethyl alcohol. This is in complete agreement with our observation.

to be larger than that of pure water(11, 12), it will be natural to conclude that its relaxation time will exceed that of free water.

The experimental results for the wet timbers cannot, of course, be treated by Eq. (1). Trying to find a more general relation than that of Debye, Cole and Cole(13) succeeded in proposing the following empirical relationship,

$$\varepsilon' - i\varepsilon'' = \varepsilon_{(0)} + \frac{\varepsilon_{(0)} - \varepsilon_{(\infty)}}{1 + (i\omega\tau)^{1-\alpha}} \qquad (5)$$

where τ is a most probable relaxation time and α is an empirical constant having a value between 0 and 1. The data for timbers are plotted in Fig. 7, as required by the relation of Cole and Cole (see Table 1).

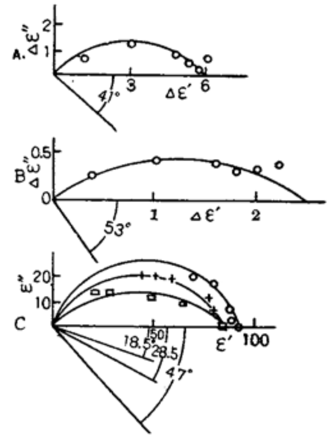

Fig. 7.—The Cole-Cole diagram.
Timber of A, 20%; B, 10% water-content. C, cane sugar solutions: ○, 30%; ×, 50%; □, 65%.

Table 1

Water-content in the sample, %	τ, $\times 10^{-10}$ sec.	α
10	1.9	0.59
20	1.3	0.45

For the comparison's sake, the values for sucrose solution(14) are also plotted in Fig. 7. The relation between the concentration and values of α in sucrose solution is similar to that of the water-content and α in the case of timber. Therefore, it may be concluded that the dielectric behaviors of wet timbers are represented by the two parameters, α and τ, with the equal accuracy to the case of sugar solutions. The values of $\Delta\varepsilon''$ measured at low frequency region do not lie on a semicircular arc in Fig. 7, this departure from the curve being attributable to the conductivity of ion.

From earlier studies of bound water, it was suggested(15, 16) that the absorbed water in timber can be grouped into three classes. (1) Strongly bound water in the region of 0—5 % water-contents, (2) weakly bound water in the region of 5—30 % water-contents — so-called capillary absorbed water, (3) nearly free water in the region of more than 30 % water-content.

The relaxation time of strongly bound water may be considered to be very large that the dielectric constant should have a small value — this being revealed by Figs. 1, 3 and 4. The fact that the rate of increase in ε becomes constant in the range of the greater water-content, *i. e.*, more than 30 %, suggests that the water of this range is nearly free in the timber.

Lastly we may add that Nukazawa and Takeda(17) have examined also some of our microwave data by the use of the theories due to Maxwell, Wagner(18), Sillars(19) and Ozawa(1, 20) for the heterogeneous dielectrics. The results appeared to be in good accord with the assumption of bound water, since we found always remarkably large loss factor and smaller dielectric constant for component water at 3000 M. C.

Summary

1. Dielectric constants and loss factors of timber *tilia* containing various amounts of water have been measured in the region of frequencies of 0.2—3000 M. C.

2. The dielectric properties of these materials at the high frequency region are found to depend upon the rotation of dipoles of water, free and bound, in timber. The empirical relation proposed by Cole and Cole are utilized for the explanation.

3. Their dielectric properties at lower frequencies may be attributed to the other causes such as the movement of ions in the material.

The present research was undertaken when the author was a member of Research Institute of Applied Electricity, Hokkaido University

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(11) Higasi, *Kagaku* (*Science*) **17**, 82, 83, 126, (1947).

(12) Higasi and Nukazawa, *Kagakuno-Kenkyu* (*Chemical Researches*) **1**, 23, (1948).

(13) Cole and Cole, *J. Chem. Phys.*, **6**, 385 (1938).

(14) Slevogt, *Ann. Phys.*, (5) **36**, 141 (1936).

(15) Stamm, *Ind. Eng. Chem.*, **21**, 94 (1929).

(16) Stamm, *J. Phys. Chem.*, **39**, 121 (1934).

(17) Nukazawa and Takeda, *Kagaku* (*Science*), **17**, 181 (1947).

(18) Wagner, *Elektrotech.*, **2**, 371 (1914).

(19) Sillars, *J. Inst. Elec. Eng.*, **80**, 378 (1937).

(20) Asami, Higasi and Ozawa, *Nature*, **165**, 195 (1950).

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