Frequency dependence of acoustic properties of aqueous glucose solutions in the VHF/UHF range

Naoyuki Akashi[a)]
Ichinoseki National College of Technology, Ichinoseki 021-8511, Japan

Jun-ichi Kushibiki
Department of Electrical Engineering, Tohoku University, Sendai 980-8579, Japan

Floyd Dunn
University of Illinois, Urbana-Champaign, 1406 West Green Street, Urbana, Illinois 61801

(Received 24 December 2003; revised 13 April 2004; accepted 29 April 2004)

The bioultrasonic spectroscopy system was employed for measurements of velocity and attenuation coefficient of glucose solutions in the VHF/UHF range. The relation between the slope of the square of velocity and the relaxation parameters, and the relation between the frequency exponent on attenuation coefficient and the relaxation parameters are investigated. In order to carry out numerical calculations, a model for a single relaxation process is employed, wherein the attenuation coefficient is expressed as $(A/(1+(f/f_\alpha)^2)+B)f^2$ where f_α is the attenuation relaxation frequency, and A and B are constants. The numerical calculations show that the slope of the square of the velocity is determined uniquely by the velocity relaxation frequency f_v and $v_\infty^2-v_0^2$ where v_0 is the zero-frequency velocity and v_∞ is the infinite-frequency velocity, and that the frequency exponent on the attenuation coefficient is determined uniquely by f_α and A/B. For experimental considerations, the velocities and the attenuation coefficients of 5, 15, and 25% concentration aqueous solutions of glucose were measured in the frequency range 20 to 700 MHz. The data for the 5 and 15% aqueous solutions can be explained using the single relaxation model. However, the data for the 25% aqueous solution suggest the existence of multirelaxation processes. [DOI: 10.1121/1.1763955]

PACS numbers: 43.80.Cs, 43.80.Ev, 43.35.Fj [WWL] Pages: 539–544

I. INTRODUCTION

The bioultrasonic spectroscopy system was developed for doing ultrasonic tissue characterization studies of biological specimens in the VHF/UHF range.[1] The acoustic properties, viz., attenuation coefficient, velocity, acoustic impedance, and density, can be determined with high accuracy by an ultrasonic transmission comparison method using pure water as the reference. The frequency dependences of the acoustic properties of bovine tissues, egg yolk and albumen, and aqueous solutions of biomacromolecules such as bovine hemoglobin and sugars measured in the frequency range have been reported.[2–4] We also have developed a measurement method for the acoustic nonlinearity parameters of liquids and biological materials in the VHF/UHF range and have been conducting basic research with the system.[5] Since the frequency characteristics of the attenuation coefficients and the velocities are obtained with high accuracy by the bioultrasonic spectroscopy, they are very useful for the research of frequency dependences of acoustic properties.[1–4] We have proposed using the exponent on the attenuation coefficient as a characterizing parameter.[1]

In this paper, the bioultrasonic spectroscopy system is employed for measurements of velocity and attenuation coefficient of aqueous glucose solutions. The relation between the slope of the square of velocity and the relaxation parameters, and the relation between the frequency exponent on the attenuation coefficient and the relaxation parameters are investigated. In order to carry out numerical calculations, a model for a single relaxation process is employed. For experimental considerations, the velocities and the attenuation coefficients of 5, 15, and 25% concentration aqueous solutions of glucose were measured in the frequency range 20 to 700 MHz and relaxation parameters were obtained.

II. NUMERICAL CALCULATIONS

In this paper, only absorption due to a relaxation phenomenon was taken into consideration as the mechanism of the propagation attenuation of ultrasonic waves. In order to carry out numerical calculations, a model for a single relaxation process is employed. Based on the acoustic relaxation theory,[6] the velocity v and the attenuation coefficient α are expressed, respectively, as

$$v^2=v_0^2+(v_\infty^2-v_0^2)\cdot\frac{(f/f_v)^2}{1+(f/f_v)^2}, \tag{1}$$

$$\alpha/f^2=\frac{A}{1+(f/f_\alpha)^2}+B, \tag{2}$$

where

[a)]Electronic mail: akashi@ichinoseki.ac.jp

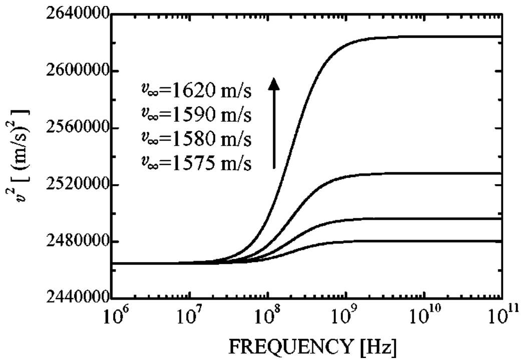

FIG. 1. Frequency dependence of the square of velocities for $v_\infty = 1575$, 1580, 1590, and 1620 m/s.

$$f_v = (v_\infty / v_0)^2 f_\alpha . \tag{3}$$

v_0 is the zero-frequency velocity and v_∞ is the infinite-frequency velocity. f is the frequency, f_v is the velocity relaxation frequency associated with the velocity, and f_α is the attenuation relaxation frequency associated with the attenuation. $A = \pi\varepsilon/(v_0 f_\alpha)$, B represents the absorption due to all other absorption mechanisms, and ε is the relaxation strength given by $\varepsilon = 1 - v_0^2/v_\infty^2$.

Numerical calculations were carried out assuming that the specimens are biological tissues or biopolymer solutions. The zero-frequency velocity v_0 was taken to be 1570 m/s. f_v in Eq. (1) was set to be 200 MHz.

Figure 1 shows the calculated results of the frequency characteristics of the square of the velocity v. In the calculations, v_∞ was taken as 1575, 1580, 1590, and 1620 m/s. v^2 has the value of v_0^2 at the zero frequency, but it gradually increases as the frequency increases. v^2 increases significantly around the velocity relaxation frequency f_v, and it has the value of v_∞^2 at the infinite frequency. It should also be noted that these curves have the inflection point at the velocity relaxation frequency f_v. The slopes of the curves in Fig. 1 are shown in Fig. 2. They are the first derivatives of v^2 with respect to the logarithm of frequency. The slope is zero at the zero frequency, but it increases as the frequency increases, and reaches its maximum value at the velocity relaxation frequency f_v. Then it decreases as the frequency increases, and becomes zero again at the infinite frequency. The maximum value of the slope is determined uniquely by the difference between v_0^2 and v_∞^2, viz., $v_\infty^2 - v_0^2$. The maximum slope increases as $v_\infty^2 - v_0^2$ increases. After all, the slope curve of the frequency characteristics of the square of the velocity v^2 is determined uniquely by f_v and $v_\infty^2 - v_0^2$. Note that when $v_\infty = 1575$, 1580, 1590, and 1620 m/s, $v_\infty^2 - v_0^2$ is 15 725, 31 500, 63 200, and 159 500 (m/s)^2, respectively.

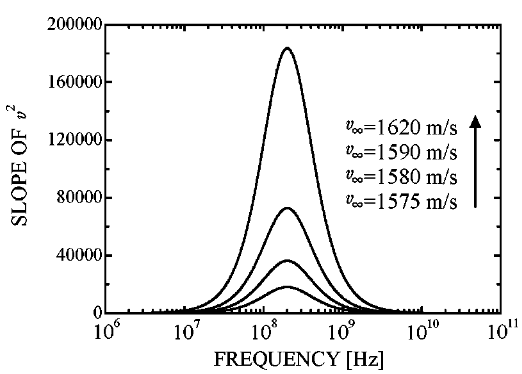

FIG. 2. Slope of the v^2 curves.

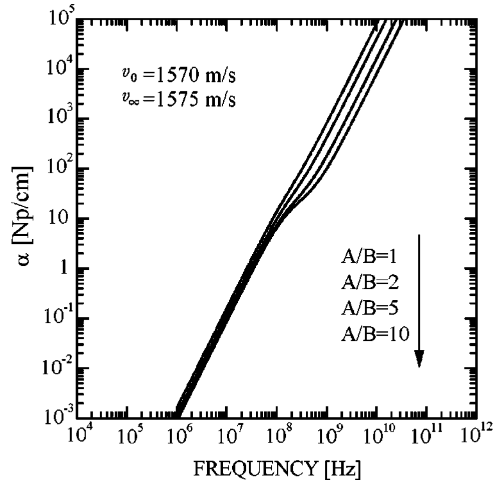

FIG. 3. Frequency dependence of attenuation coefficients for $A/B = 1$, 2, 5, and 10.

Figure 3 shows the calculated results of the frequency characteristics of the attenuation coefficient α. In the calculations, $v_0 = 1570$ m/s, $v_\infty = 1575$ m/s, and B was set to satisfy $A/B = 1$, 2, 5, and 10. The attenuation relaxation frequency f_α is calculated to be 198.7 MHz from Eq. (3). For every A/B, the slope of the α curve is 2 at the zero frequency, but it decreases around the attenuation relaxation frequency f_α, and becomes 2 again at the infinite frequency. Figure 4 shows the frequency characteristics of the exponent on the attenuation coefficient α in Fig. 3. The exponent on α is 2 both at the zero and infinite frequencies, but it decreases significantly in the neighborhood of the attenuation relaxation frequency f_α and reaches its minimum value. It is found that the minimum value of the exponent on the attenuation coefficient decreases as A/B increases. It is also observed that the frequency, at which the exponent on the attenuation coefficient reaches its minimum, increases as A/B increases, even though the attenuation relaxation frequency f_α is unchanged.

It is concluded that the frequency characteristics of the exponent on the attenuation coefficient are determined

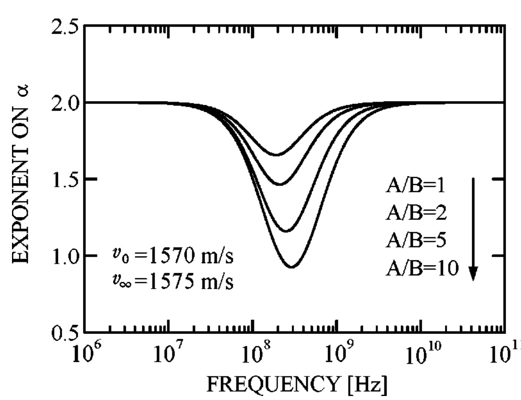

FIG. 4. Frequency dependence of the exponent on attenuation coefficients for $A/B = 1$, 2, 5, and 10.

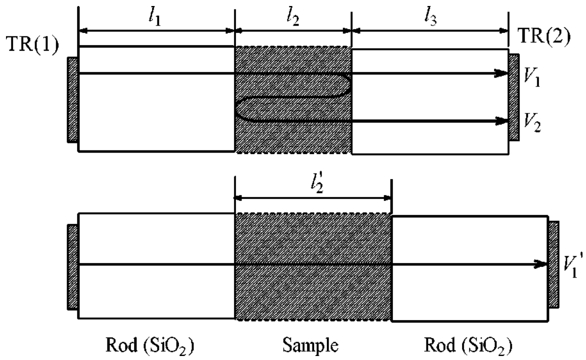

FIG. 5. Definition of transducer outputs V_i for measuring acoustic properties of biological media.

uniquely by the attenuation relaxation frequency f_α and the relaxation parameter ratio A/B. Note that scattering in the media due to structural features may affect the exponent on the attenuation coefficient, for example, in biological media.

III. MEASUREMENT METHOD

Measurements were performed using the bioultrasonic spectroscopy system. The system is capable of measuring acoustic properties, viz., attenuation coefficient, velocity, acoustic impedance and density of liquid, biological tissues, and solids in the VHF/UHF range. The system is described in detail elsewhere.[1] The system has further been improved to measure the amplitude and phase of radio frequency (rf) tone burst signals in the VHF range.[7,8]

The experimental configuration for measurements is illustrated schematically in Fig. 5, in which a specimen is inserted between the parallel surfaces of synthetic silica (SiO_2) buffer rods having ZnO piezoelectric film transducers on their outer ends. The gap length between the two buffer rods is determined by measuring frequency characteristics of the interference output of V_1 and V_2 for pure water of which the velocity is used as the reference.[1]

The velocity is obtained by the complex-mode velocity measurement method,[7,8] in which the phase of the transducer output V_1 and V_2 is measured as a function of frequency. From the measured phase difference $\phi\,(0<\phi<2\,\pi)$ between V_1 and V_2, the velocity is obtained as

$$v=-\frac{4\pi f l_2}{(\phi+2\pi n)-\Delta\theta}, \tag{4}$$

where n is the integer number and $\Delta\theta$ is the phase advance effect due to diffraction, which can be corrected numerically. The numerical calculations are performed using the exact integral expression of diffraction by Williams.[9] Since the value of the measured phase difference ϕ is smaller than 2 π, the value of n must be determined. If the velocity at a frequency is given, n can be determined by using Eq. (4). The velocity was measured at one frequency using the z-interference method,[1] in which the interference output of V_1 and the reference electrical signal derived from the signal generator of the measurement system is measured as a function of gap length l_2.

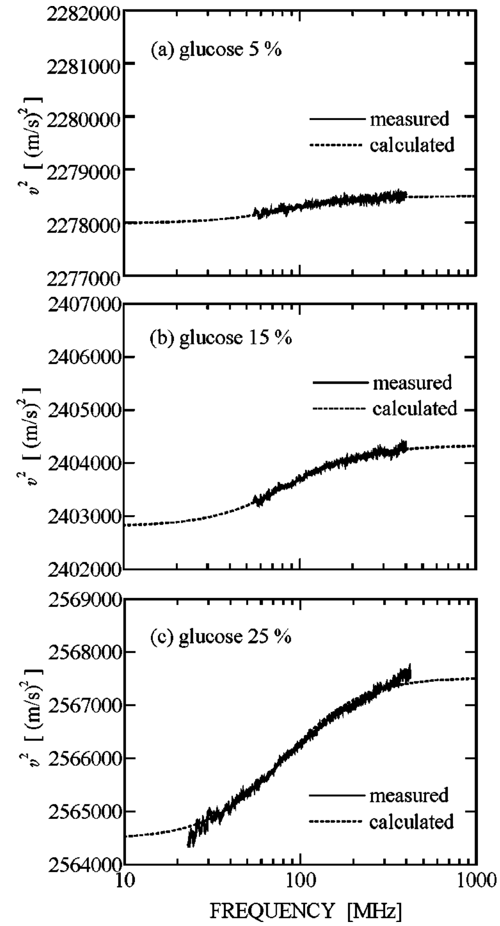

FIG. 6. Measured and calculated frequency dependence of the square of velocities of 5, 15, and 25% aqueous solutions of glucose at 23 °C.

The attenuation coefficient is determined by measuring the amplitude of the transducer outputs V_1 and V_1' for different gap lengths l_2 and l_2' as

$$\alpha=\frac{1}{l_2'-l_2}\ln\frac{|V_1|}{|V_1'|}-\frac{1}{l_2'-l_2}\cdot\ln\frac{|ATT_1|}{|ATT_1'|}, \tag{5}$$

where ATT_1 and ATT_1' are the diffraction loss for the transducer outputs V_1 and V_1', respectively. The diffraction loss can be corrected numerically using the exact integral expression of diffraction by Williams.[9]

IV. EXPERIMENTS AND RESULTS

5, 15, and 25% concentration aqueous solutions of glucose were used. The frequency characteristics of the velocities and the attenuation coefficients were measured using the bioultrasonic spectroscopy system. To cover the frequency range from 20 to 700 MHz, four different ultrasonic devices having operating center frequencies of 70, 150, 420, and 700 MHz, respectively, were employed. The temperature of the specimen was controlled to be $23.00\pm0.05\,°C$.

A. Velocity

Figure 6 shows the measured frequency characteristics of the square of the velocity v^2 for 5, 15, and 25% aqueous solutions of glucose in the frequency ranges 55 to 400 MHz, 55 to 400 MHz, and 23 to 420 MHz, respectively. It is seen

TABLE I. Acoustic parameters used for numerical calculations of the velocities and the attenuation coefficients.

		Concentration		
		5%	15%	25%
Velocity measurement	v_0 [m/s]	1509.30	1550.10	1601.40
	v_∞ [m/s]	1509.47	1550.59	1602.35
	f_v [MHz]	85	85	81
	$A[\times 10^{-17}\ \mathrm{s^2/cm}]$ (determined using v_0, v_∞, and f_v)	5.5	15.1	28.7
Attenuation measurement	$A[\times 10^{-17}\ \mathrm{s^2/cm}]$	6.0	15.0	29.0
	$B[\times 10^{-17}\ \mathrm{s^2/cm}]$	23.5	27.0	32.5
	f_α [MHz]	85	85	84

that the velocities increase with frequency for every concentration. The dotted curves in the figures are the calculated results of v^2 with the acoustic parameters of v_0, v_∞, and f_v listed in Table I. Figure 7 shows the determined slopes of the frequency characteristics of v^2 in Fig. 6. The maximum value of the slope increases with concentration. The dotted curves in the figures are the calculated results. The measured curves are in excellent agreement with the calculated curves in both Figs. 6 and 7 for the concentration of 5 and 15%. However, there are differences in shape between the measured and calculated velocity curves for the 25% aqueous solution. By comparing Figs. 6 with 7, it is seen that the velocity relaxation frequency f_v is clearly detected in the frequency characteristics of the slopes of v^2. From the frequency characteristics of the slopes of v^2 shown in Fig. 7, f_v and $v_\infty^2 - v_0^2$ can be determined. $v_\infty^2 - v_0^2$ was determined as 513, 1519, and 3044 $(\mathrm{m/s})^2$ for the concentration of 5, 15, and 25%, respectively.

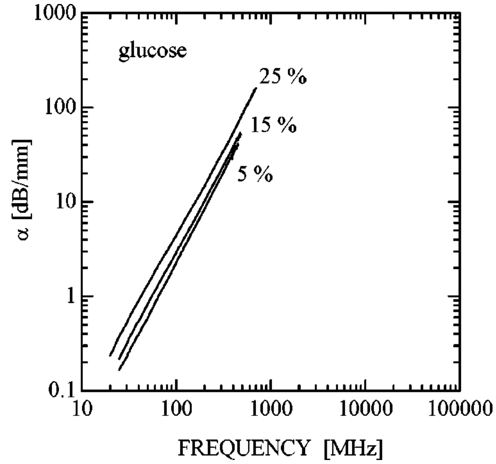

FIG. 8. Measured attenuation coefficients α of 5, 15, and 25% aqueous solutions of glucose at 23 °C.

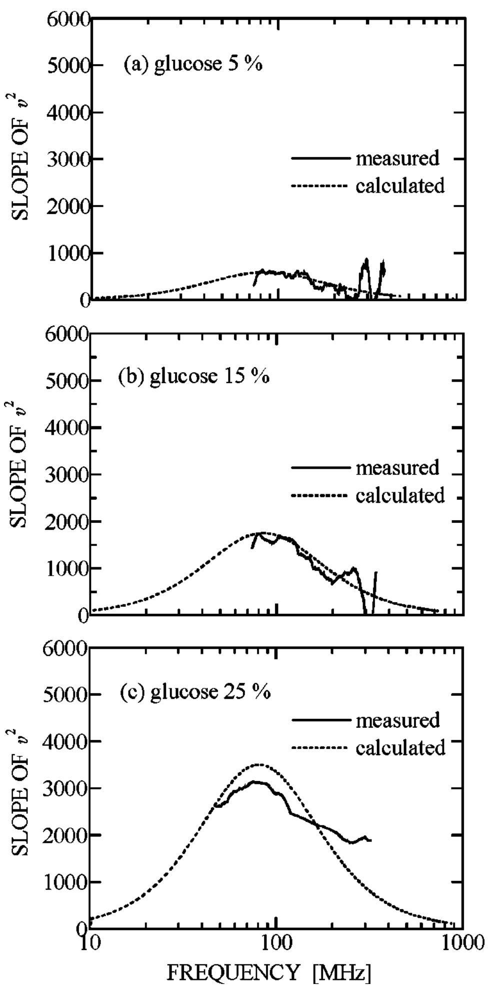

FIG. 7. Measured and calculated frequency dependence of the slope of the square of velocities of 5, 15, and 25% aqueous solutions of glucose at 23 °C.

B. Attenuation coefficient

Figure 8 shows the measured frequency characteristics of the attenuation coefficients α for 5, 15, and 25% aqueous solutions of glucose in the frequency ranges 25 to 450 MHz, 25 to 480 MHz, and 20 to 700 MHz, respectively. Figure 9 shows the attenuation coefficient α divided by the square of frequency f, viz., α/f^2. The solid and dotted curves represent the measured and calculated values, respectively. The calculations were performed using Eq. (2) with the acoustic parameters of A, B, and f_α listed in Table I. Figure 10 shows the determined frequency characteristics of the exponent on α in Fig. 8. The solid and dotted curves in the figure represent the measured and calculated values, respectively. The measured curves are in agreement with the calculated curves in both Figs. 9 and 10. From the exponent on α shown in Fig. 10, f_α and A/B can be determined. A/B was determined as 0.255, 0.556, and 0.892 for the concentration of 5, 15, and 25%, respectively.

C. Discussion

The value of A in Eq. (2) can be determined either by the velocity or the attenuation measurement. According to the relations of $A = \pi\varepsilon/(v_0 f_\alpha)$, $\varepsilon = 1 - v_0^2/v_\infty^2$, and $f_v = (v_\infty/v_0)^2 f_\alpha$, A is determined using the values of v_0, v_∞, and f_v. Table I shows the values of A determined by the velocity and attenuation measurements, where it is seen that they are in excellent agreement with each other for the concentrations of aqueous solutions of glucose.

For the 5 and 15% aqueous solutions of glucose, the measured curves are in excellent agreement with the calculated curves for both velocity and attenuation coefficient.

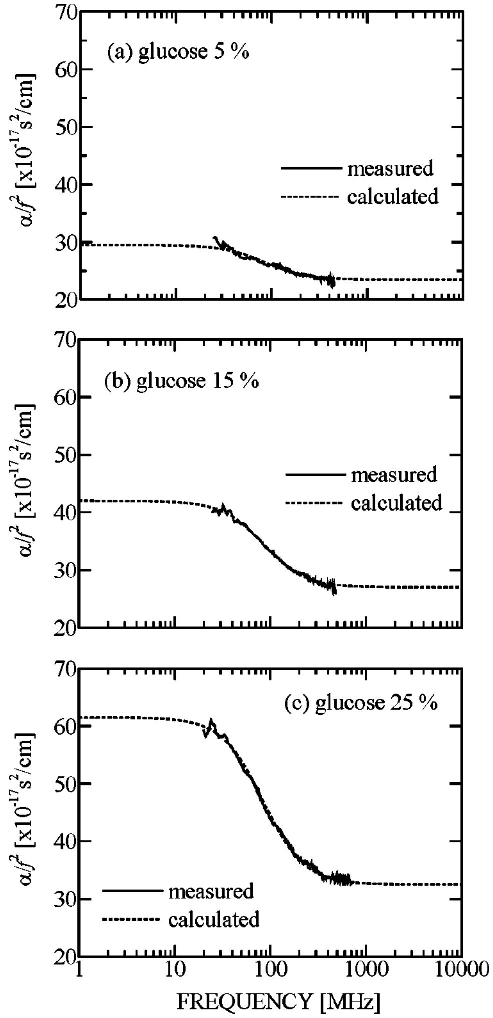

FIG. 9. Measured and calculated α/f^2 of 5, 15, and 25% aqueous solutions of glucose at 23 °C.

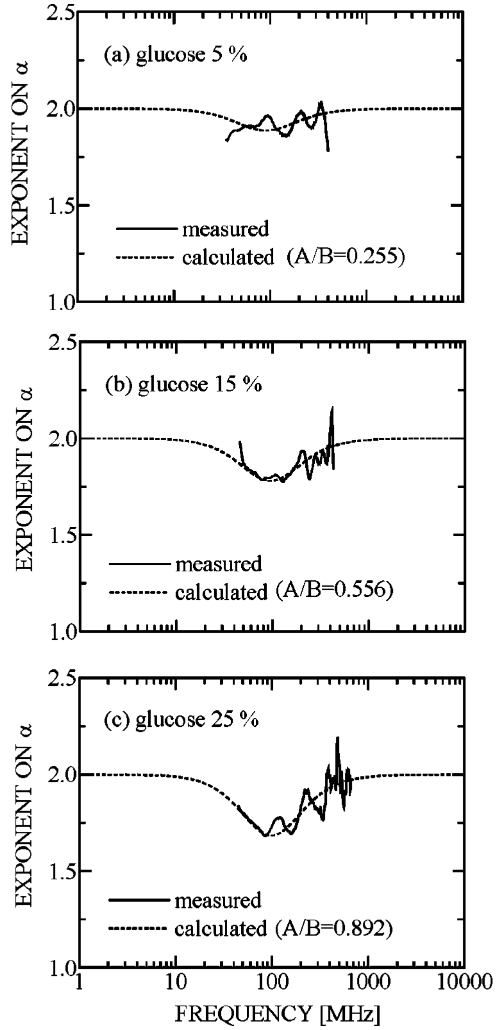

FIG. 10. Measured and calculated frequency dependence of the exponent on attenuation coefficients of 5, 15, and 25% aqueous solutions of glucose at 23 °C.

However, there are differences in shape between the measured and calculated velocity curves for the 25% aqueous solution. Further, $f_v = 81$ MHz was obtained from the velocity, and $f_\alpha = 84$ MHz was obtained from the attenuation coefficient for the 25% aqueous solution. The difference between them is 4%. Since $f_v = 1.0012 f_\alpha$ is obtained by substituting $v_0 = 1601.40$ m/s and $v_\infty = 1602.35$ m/s into Eq. (3), viz., $f_v = (v_\infty / v_0)^2 f_\alpha$, the difference between f_v and f_α which is predicted from the difference between v_0 and v_∞ is 0.12%. The causes of the 4% difference between the measured values of f_v and f_α cannot be explained by this relaxation theory. It is necessary to analyze the data by introducing multirelaxation processes. Since the value of the relaxation frequencies obtained from the 5, 15, and 25% aqueous solutions are nearly equal, a main relaxation process may be due to the same mechanism for all the concentration solutions. A second relaxation process in the 25% aqueous solution may arise from solute–solute interaction processes.

V. SUMMARY

In this paper, the bioultrasonic spectroscopy system has been employed for measurements of velocity and attenuation coefficient of aqueous glucose solutions in the VHF/UHF range. The relation between the slope of the square of velocity and the relaxation parameters, and the relation between the frequency exponent on attenuation coefficient and the relaxation parameters were numerically calculated and presented. It was shown that the slope of the frequency characteristics of the square of the velocities was determined uniquely by the velocity relaxation frequency f_v, and the difference between the square of the velocity at zero frequency v_0^2 and the square of the velocity at infinite frequency v_∞^2, viz., $v_\infty^2 - v_0^2$. Also, the exponent on the attenuation coefficient α was determined uniquely by the attenuation relaxation frequency f_α and the ratio of the parameters A/B, when $\alpha/f^2 = A/(1+(f/f_\alpha)^2) + B$.

To verify the calculated results through experiments, the frequency characteristics of velocities and attenuation coefficients were measured for 5, 15, and 25% concentration aqueous solutions of glucose in the frequency range from 20 to 700 MHz. The measured data for the 5 and 15% aqueous solutions can be explained using the single relaxation model. On the other hand, it is necessary to introduce a model for multirelaxation processes in order to understand the data for the 25% aqueous solution.

[1] J. Kushibiki, N. Akashi, T. Sannomiya, N. Chubachi, and F. Dunn, ''VHF/UHF range bioultrasonic spectroscopy system and method,'' IEEE Trans. Ultrason. Ferroelectr. Freq. Control **42**, 1028–1039 (1995).
[2] N. Akashi, J. Kushibiki, N. Chubachi, and F. Dunn, ''Acoustic properties of selected bovine tissues in the frequency range 20 to 200MHz,'' J. Acoust. Soc. Am. **98**, 3035–3039 (1995).
[3] N. Akashi, J. Kushibiki, and F. Dunn, ''Acoustic properties of egg yolk and albumen in the frequency range 20 to 400 MHz,'' J. Acoust. Soc. Am. **102**, 3774–3778 (1997).
[4] N. Akashi, J. Kushibiki, and F. Dunn, ''Measurements of acoustic properties of aqueous dextran solutions in the VHF/UHF range,'' Ultrasonics **38**, 915–919 (2000).
[5] J. Kushibiki, M. Ishibashi, N. Akashi, T. Sannomiya, N. Chubachi, and F. Dunn, ''Transmission line method for the measurement of the acoustic nonlinearity parameter in biological liquids at very high frequencies,'' J. Acoust. Soc. Am. **102**, 3038–3044 (1997).
[6] A. B. Bhatia, ''Discussion of experimental data in polyatomic liquids,'' in *Ultrasonic Absorption* (Dover, New York, 1967), Chap. 10, pp. 194–263.
[7] J. Kushibiki and M. Arakawa, ''Diffraction effects on bulk-wave ultrasonic velocity and attenuation measurements,'' J. Acoust. Soc. Am. **108**, 564–573 (2000).
[8] J. Kushibiki, R. Okabe, and M. Arakawa, ''Precise measurements of bulk-wave ultrasonic velocity dispersion and attenuation in solid materials in the VHF range,'' J. Acoust. Soc. Am. **113**, 3171–3178 (2003).
[9] A. O. Williams, Jr., ''The piston source at high frequencies,'' J. Acoust. Soc. Am. **23**, 1–6 (1951).