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Design of Low-Loss 1–3 Piezoelectric Composites for High-Power Transducer Applications

Hyeong Jae Lee and Shujun Zhang [Senior Member, IEEE]

Materials Research Institute, The Pennsylvania State University, University Park, PA (soz1@psu.edu).

Abstract

Lead zirconate titanate (PZT)/polymer 1–3 composites have improved electromechanical properties compared with monolithic counterparts, but possess a low mechanical quality factor, limiting their use in high-power transducer applications. The goal of this work was to improve the mechanical quality factor of 1–3 PZT/polymer composites by optimizing the polymer materials. Theoretical analysis and modeling were performed for optimum composite design and various polymers were prepared and characterized. 1–3 piezocomposites were constructed and their electromechanical properties were experimentally determined. The results demonstrated that the composites with high-thermal-conductivity polymers generally have degraded electromechanical properties with significantly decreased mechanical quality factors, whereas the composites filled with low-loss and low-moduli polymers were found to have higher mechanical quality factors with higher electromechanical coupling factors: $Q_m \sim 200$ and $k_t \sim 0.68$ for PZT4 composites; $Q_m \sim 400$ and $k_t \sim 0.6$ for PZT8 composites. The improved mechanical quality factor of 1–3 piezocomposites may offer improved performance and thermal stability of transducers under high-drive operation.

I. Introduction

PIEZOELECTRIC polymer composites with 1–3 connectivity consist of length-extensional piezoelectric elements embedded in a passive matrix. The benefits of 1–3 piezocomposites are well-recognized for transducer imaging applications, taking advantages of the high electromechanical properties of piezoelectrics and low acoustic impedance of polymers. One principal advantage is that this structure allows the use of the length-extensional coupling of piezoelectrics for thickness-mode transducers, enabling broader bandwidth and higher sensitivity; therefore, they have been widely used in medical diagnostics and nondestructive testing transducers [1]–[3].

Ultrasonic transducers for high-power applications, such as high-intensity focused ultrasound and underwater sonar projectors, are generally operated in a frequency band around the fundamental resonance to maximize output power. The acoustic output power (P) at resonance can be defined as the input electrical power (U_e) reduced by the electroacoustic efficiency (η_{ea}) of the transducer, i.e., $P = \omega Q_m \eta_{ea} U_e$, and the efficiency can be determined from the product of electromechanical efficiency and mechanoacoustical efficiency. The electromechanical efficiency is the ratio of the power delivered to the mechanical part of the transducer to the input electrical power, which is the definition of the electromechanical coupling factor. The mechano-acoustical efficiency is the ratio of acoustical power radiated into the medium to the total power delivered to the mechanical and acoustical sections, given by:

$$\eta_{\text{ma}} = \frac{R_r}{(R_r + R_m)}, \quad (1)$$

where R_r is the radiation resistance and R_m is the internal mechanical resistance, which is related to the mechanical loss, indicating the importance of electromechanical coupling factor and mechanical quality factor Q_m (inverse of mechanical loss) of piezoelectric materials for high-power applications [4], [5].

Another important consideration for high-power applications is power dissipation in the form of heat, as a result of a rise in the mechanical loss under high-drive operation. This limits the maximum allowable driving field and, therefore, output power of the transducers [6], [7]. Conventional 1–3 piezocomposites are not suitable for high-power transducers because of the high mechanical loss resulting from the presence of the polymer matrix. Furthermore, the polymer phase exhibits a low thermal conductivity (0.2 to 0.3 W/m·K), hindering heat dissipation during high-drive operation [8]–[10]. Therefore, to improve the high-power characteristics of composite transducers, it is necessary to minimize the losses in a piezocomposite or to improve the thermal conductivity of polymer matrix to allow higher acoustic output power without excessive heat generation.

The objective of this research is the design of high-mechanical-quality-factor 1–3 piezocomposites for high-power transducer applications. In this work, optimization of the composite design and properties of its constituents were theoretically and experimentally investigated. Electrical impedance measurements were conducted on various piezocomposites with different types of PZT ceramics and polymers to assess their electromechanical properties and mechanical quality factors. The effect of composite filler material on the performance of composites was discussed to provide appropriate composite filler materials for high-power applications.

II. Experimental and Theoretical Methods

A. Selection of Composite Components

The primary component of the composites is the selection of the piezoelectric material, which determines the maximum electromechanical coupling factor and mechanical quality factor. In this study, DoD I and III type lead zirconate titanate ceramics (PZT: PKI406 and PKI809, Piezokinetics, Bellefonte, PA) were selected, offering low losses ($\tan \delta < 0.004$, $Q_m > 500$) with relatively high electromechanical coupling factors ($k_{33} = 0.64$ to 0.7), and their suitability has been demonstrated for a wide range of high-power applications [11], [12].

For the passive phase, commercially available thermosetting epoxy polymers were evaluated. The two most important parameters of the polymers for high-power composites are the elastic stiffness and loss that is related to viscosity; both of which contribute to the mechanical quality factor. Thermal conductivity is also an important material parameter because low thermal conductivity limits heat transfer, degrading thermal management of transducer under high-drive operation. In this study, two different groups of polymers were prepared and characterized for composite fillers. One group is low-elastic-stiffness and low-loss polymers, specifically (I) Spurr resin (Polyscience Inc., Warrington, PA) and (II) Poss EP3512 resin (Hybrid Plastics Inc., Hattiesburg, MS); the other group includes high-thermal-conductivity polymers (>1 W/m·K), which were (A) T7110 (Epoxy Technology Inc., Billerica, MA), (B) Duralco 128 (Cotronics Corp., Brooklyn, NY) and (C) Stycast 2850FT (Emerson & Cuming Specialty Polymers, Billerica, MA).

B. Composite Design and Fabrication

In designing a 1–3 piezocomposite, it is important to avoid coupling of spurious modes with the desired thickness vibration, because mode coupling reduces the transmitting energy and consequently reduces transducer performance. For consideration of piezoelectric elements inside 1–3 piezocomposites, the half-wave lateral mode of the elements should be well above the fundamental thickness vibration so that their harmonics do not couple to the thickness mode. This requires a low width-to-thickness aspect ratio of the piezoelectric elements [13], [14]. Fig. 1 shows examples of impedance and phase angle as a function of frequency at various width-to-thickness aspect ratios (G) of PZT ceramics. It can be observed that as the aspect ratio increases, the thickness mode vibration was significantly disturbed by other vibration modes, resulting in considerable degradation of electromechanical coupling factor.

Because of the periodic structure of 1–3 piezocomposites, there are laterally propagating Lamb waves and the interference of these resonances with the thickness resonances also degrades transducer performance. The first stopband edge resonance occurs when the wavelength of Lamb waves approaches the half-wavelength of the shear wave of the filler. Therefore, the spacing (s) between piezoelectric elements should be less than $v_s/(2f)$, where v_s and f are the shear wave velocity of the polymer fillers and the thickness resonant frequency of the composites, respectively. This condition places the stopband edge resonant frequencies above the thickness resonant frequency. For the same reason, the width (w) of the piezoelectric elements should be less than $v_L/(2f)$, where v_L is the lateral wave velocity of a piezoelectric element. The general guideline for the spatial scale of a 1–3 composite is the stopband edge resonant frequency should be located more than two times the fundamental thickness resonant frequency, which gives the following conditions for the composite design [15], [16]:

$$s < \frac{v_s}{4f}, \quad \text{and} \quad w < \frac{v_L}{4f}. \quad (2)$$

Based on these conditions, composites were fabricated using the conventional dice-and-fill technique. To avoid the frequency-dependent attenuation property variation of each polymer, the resonant frequency of all fabricated composites was kept constant at ~400 kHz, and the width of the piezoelectric elements and spacing between elements were determined based on the conditions in (2). A 100-mm-diameter, 0.3-mm-thick diamond wafering blade was used in this study, which gives a kerf width (spacing) of approximately 0.3 mm. The samples were first diced along one direction, and then in the perpendicular direction. A range of polymers were then backfilled into the kerfs under a vacuum, and subsequently cured according to the manufacturer's instructions. The fabricated composites were parallel polished until all of the piezoelectric posts were exposed.

C. Characterization Methods

The elastic properties and attenuation coefficients of the thermosetting polymers were characterized by using a through-transmission technique. The transducers, a 10-MHz longitudinal wave transducer and a 5-MHz shear wave transducer, were excited by a 200-MHz computer-controlled pulser (5900 PR, Panametrics NDT Inc., Waltham, MA), and the travel time and amplitude variance between echoes were measured using a digital oscilloscope (TDS 460A, Tektronix Inc., Beaverton, OR). This allowed for the calculation of the longitudinal (v_L) and shear (v_s) velocities and the attenuation coefficient (α). Elastic constants (c_{ij}) were then determined from the Christoffel wave equation, which is the relationship between the phase velocities and elastic constants of the material:

$$c_{11}=\rho v_L^2, \quad c_{44}=\rho v_s^2, \quad c_{12}=c_{11}-2c_{44}, \quad (3)$$

and the Poisson's ratio (σ) of polymers was derived from the obtained elastic constants according to

$$\sigma=\frac{c_{12}}{2(c_{12}+c_{44})}. \quad (4)$$

The thermal conductivity (k) of polymers was calculated by measuring the density (ρ), thermal diffusivity (α_d) and heat capacity (C_p) using a Netzsch LFA457 microflash (Netzsch-Geratebau, Selb, Germany):

$$k=\rho\alpha_d C_p. \quad (5)$$

The glass transition temperature or maximum thermal degradation temperature of polymers was determined by measuring the dielectric loss as a function of temperature, because an abrupt rise in loss is generally associated with the glass transition temperature [17].

The electromechanical coupling factor (k_t) and mechanical quality factor (Q_m) of the 1–3 piezocomposites were calculated from the measured resonant frequencies and equivalent circuit parameters, which were measured with an HP4194A impedance/gain-phase analyzer (Hewlett-Packard, Palo Alto, CA):

$$k_t=\frac{\pi}{2}\frac{f_r}{f_a}\cot\left(\frac{\pi}{2}\frac{f_r}{f_a}\right), \quad (6)$$

$$Q_m=\frac{1}{\omega RC_m}, \quad (7)$$

where f_r and f_a are the resonant and anti-resonant frequencies, respectively. R and C_m are the resistance and capacitance of the series branch of the Butterworth–Van Dyke equivalent circuit. The mechanical quality factor of composites was also determined from the attenuation

$$Q_m=\frac{\beta}{2\alpha}, \quad (8)$$

which is based on the transmission-line model, in which the attenuation coefficient of piezocomposites can be determined by [18]

$$\alpha=\frac{1}{l}\ln\left(\frac{(D+1)}{(D-1)}\right), \quad (9)$$

where β is the wave number and $D=(\pi^2 f_a R_M C^s)/k_t^2$, where R_M and C^s are the resistance at f_a and clamped capacitance, respectively. The benefit of this method is that mechanical quality factor is less influenced by the electrode impedance compared with the method in (7) because R_M reaches a maximum value at f_a .

III. Results and Discussion

A. Single-Phase Polymers

Table I summarizes the properties of various polymer materials. From the table, it can be observed that there is a tradeoff between elastic stiffness and thermal conductivity; polymers (I and II) have low loss determined by attenuation and stiffness but low thermal conductivity, whereas polymers (A–C) have high thermal conductivity, but high loss and stiffness. These tradeoffs arise from the fact that high thermal conductivity is generally achieved by the addition of thermally conductive filler particles, such as AlN and BN [19], [20]. These fillers result in increased elastic stiffness and attenuation of the polymer composites.

Fig. 2 shows the temperature-dependent dielectric loss tangent ($\tan \delta$) of the various polymers, demonstrating the thermal stability of each polymer. From the figure, the polymers (I) and (C) did not show an observable glass transition and are able to retain the dielectric properties at elevated temperature up to 180°C, whereas those of polymers (II) and (B) increased rapidly above 60°C as a result of polymer softening. Such low-glass-transition-temperature polymers have greatly limited use in high-power applications in which temperature rises with increasing driving field. Polymer (A) showed relatively higher thermal stability than polymers (II) and (B); however, the dielectric loss gradually increased above 100°C, limiting its use above 100°C.

B. 1–3 Piezocomposites

The model developed by Smith and Auld [21] has provided a prediction of the electromechanical properties of 1–3 piezocomposites, offering useful guidelines in the composite design. This modeling technique was used to predict the performance of 1–3 piezocomposites before the experimental confirmation. The interesting material parameters from this modeling are electromechanical coupling factor and wave velocity of a 1–3 piezocomposite; the coupling factor reflects the performance of a 1–3 piezocomposite as a transducer and the wave velocity allows for the estimation of resonant frequency for a given composite thickness. The following equations were used to calculate the coupling factor and velocity of 1–3 composites:

$$k_t^* = \frac{\overline{e_{33}}}{\sqrt{\overline{c_{33}^D} \overline{\epsilon_{33}^S}}}, \quad v_t^* = \sqrt{\frac{\overline{c_{33}^D}}{\overline{\rho}}}, \quad (10)$$

with

$$\begin{aligned} \overline{e_{33}} &= v(e_{33} - 2e_{31}(c_{13}^E - c_{12})/c(v)), \\ \overline{c_{33}^D} &= c_{33}^E + e_{33}^2/\epsilon_{33}^S, \\ \overline{c_{33}^E} &= v(c_{33}^E - 2(c_{13}^E - c_{12})/c(v)) + (1-v)c_{11}, \end{aligned}$$

where the overline denotes a composite parameter, v is the ceramic volume fraction, and $\alpha(v) = c_{11}^E + c_{12}^E + [v(c_{11} + c_{12})/(1-v)]$. ϵ_{33}^S is approximated to $\sim v\epsilon_{33}^S$ because of the much higher ϵ_{33}^S of the piezoelectric ceramic. $\overline{\rho}$ is equal to $v\rho_c + (1-v)\rho_p$ where ρ_c and ρ_p are the densities of the ceramic and polymer, respectively.

Modeling results of electromechanical coupling factor and wave velocity of the various 1–3 piezocomposites as a function of volume fraction are given in Fig. 3. From the modeling,

PZT4 ceramics were selected as a piezoelectric material whose material parameters are listed in Table II. The modeled results of PZT8 composites are not included here because the behaviors of the PZT8 composites are similar to those of PZT4 composites. In general, polymers (I) and (II) offered higher electromechanical properties compared with high thermal conductivity polymers (A)–(C), which can be explained by the relatively high elastic moduli of polymers (A)–(C), which tends to clamp the vibration of the piezoelectric elements. The wave velocity of composites decreased with decreasing volume fraction, and the composites with polymers (I) and (II) showed lower velocity compared with those with polymers (A)–(C) as a result of lower elastic modulus of polymers (I) and (II).

Fig. 4 shows the experimentally determined coupling and mechanical quality factor of 1–3 polymer (I)/PZT4 composites as a function of volume fraction. In general, good agreement was found between the theoretical and experimental values for the coupling factor. It can be seen that the coupling factors of composites using polymer (I) were found to be 0.65 to 0.68 over the range of volume fraction, similar to k_{33} values of monolithic ceramics. The mechanical quality factor of composites showed much lower values compared with monolithic PZT4 ($Q_m \sim 500$), and exhibited a decreasing trend with decreasing piezoelectric volume fraction, being in the range of 150 to 200.

Based on the modeling and experimental results, 1–3 piezocomposites with a PZT volume fraction of 0.70 were fabricated because they had the highest electromechanical coupling and mechanical quality factors.

Table III summarizes the material properties for the fabricated 1–3 piezocomposites depending on the polymer material. For coupling factors, 1–3 composites using polymers (I) and (II) exhibited values similar to the k_{33} values in monolithic PZT samples, being on the order of 0.68 for PZT4 composites and 0.60 for PZT8 composites. For the case of high-thermal-conductivity polymers, there were several issues when they were used in composite filler materials. For example, high-thermal-conductivity polymers generally have high viscosity in the uncured state, which makes it difficult to fill in the kerf during composite fabrication, especially for the polymer (C). In addition, the polymers have relatively high moduli compared with polymers (I) and (II), resulting in reduced coupling factors. However, it should be noted that all 1–3 piezocomposites exhibited higher coupling than the thickness coupling in monolithic PZTs ($k_t = 0.45$ to 0.50), confirming the advantage of 1–3 piezocomposites over their monolithic counterparts.

For the case of mechanical quality factor Q_m , it should be noted that mechanical Q_m of the composites determined from (8) generally show higher values than those determined using the equivalent circuit model (IEEE method). The possible measurement variations from these methods are due to the accuracy of measuring the minimum impedance, R , at resonant frequency for IEEE method, and the R_M , C^S , and k_t for transmission line model.

From Table III, it can be seen that the mechanical quality factors of the composites were found to decrease significantly compared with those of monolithic bulk materials, especially those filled with high-thermal-conductivity polymers ($Q_m < 100$). However, it is interesting to note that the mechanical Q_m of the composites with polymer (I) is considerably higher than those of other composites, being on the order of 200 for PZT4 composites and 400 for PZT8 composites, making them promising materials for high-power applications. The reasons for the relatively low mechanical quality factor of 1–3 composites with high-thermal-conductivity polymers (A)–(C) can be explained by the contribution of the inherent damping of the piezo-electrics from the surrounding polymer, which increases with increasing elastic stiffness of the polymer. Therefore, the polymers (A)–(C) not only

decrease electromechanical coupling factor but also decrease mechanical quality factor of 1–3 piezocomposites because of their higher elastic stiffness.

In addition to the polymer's stiffness, another important contribution to mechanical quality factor of composites is the elastic loss of polymers. For example, the elastic loss of polymers is expressed as the ratio of the imaginary and real parts of the elastic constant [17]. In wave phenomena, this is generally described by the attenuation, as indicated in (8). The approximation of mechanical wave attenuation of 1–3 piezocomposites is given by (11) [22]:

$$\alpha = v\alpha_p + (1 - v)\alpha_e, \quad (11)$$

where v is the volume fraction and the subscripts p and e refer to piezoelectric and epoxy polymer, respectively. This indicates that the mechanical quality factor of composites is strongly affected by the elastic loss of polymers (see Table I).

Thermal stability of 1–3 piezocomposites with different filler materials was further investigated at elevated temperature. Note that the temperature-dependent properties of PZT8 composites were not included here because the behavior of PZT8 composites with temperature is similar to that of PZT4 composites. Because of their higher coupling and mechanical quality factors, only composites (I) and composites (A) were investigated in this study. Fig. 5 shows the change in resonant frequency and mechanical quality factor of PZT4 composites (I) and (A) as a function of temperature. It was shown that as the temperature increases, both composites showed a decrease in resonant frequency, which is a consequence of a reduction in velocities, because the elastic moduli of the constituents decrease with increasing temperature. However, compared with polymer (I) composites, polymer (A) composites showed a larger decrease in resonant frequency, which is reflected by the influence of different glass transition temperature of composite fillers, as shown in Fig. 2. The mechanical quality factor of 1–3 piezocomposites increased with increasing temperature, indicating that the epoxies softened with increasing temperature, thus leading to less clamping effect on piezoelectric component.

IV. Conclusion

This work has demonstrated that the electromechanical coupling and mechanical quality factor of 1–3 piezocomposites can be improved with the appropriate selection of polymer. The optimum design of 1–3 composites was determined based on the unidimensional modeling, and the properties of 1–3 composites were experimentally determined with particular emphasis on electromechanical coupling and mechanical quality factor. The results indicated that the composites with low-loss and low-moduli polymer (I) offered relatively high mechanical quality factor, being on the order of ~200 and ~400 for PZT4 and PZT8 composites. In addition, the polymer (I) composite also offered an improved electromechanical coupling factor compared with other polymer fillers, approaching k_{33} values of monolithic PZTs, being 0.68 for PZT4 composites and 0.60 for PZT8 composites. In contrast, although the use of high-thermal-conductivity polymers can provide the potential to improve thermal stability of 1–3 piezocomposites, because they act as an effective thermal pathway to the load medium, these polymers generally have high elastic moduli and high attenuations, resulting in a significant reduction in mechanical quality factor, $Q_m < 100$, as well as a decrease in coupling factor.

Further investigations of 1–3 piezocomposites under high-drive conditions will be carried out, such as field dependence of maximum vibration velocity and mechanical quality factor, and duty cycle dependence of maximum power level without exceeding a predetermined

temperature. In addition, the performance of composites with low-loss polymer and high-thermal-conductivity polymer will be compared under high-drive conditions.

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Biographies

Hyeong Jae Lee was born in South Korea and received his B.S. and M.S. degrees in materials science and engineering from The Pennsylvania State University, University Park, PA. He is currently a Ph.D. candidate at the Material Research Institute of The Pennsylvania State University.

His research interests include ceramic processing, material characterization, high-frequency broadband ultrasonic transducers, and high-power ultrasonic transducers. He is particularly interested in the study of materials that address the fundamental relations between microstructure, processing, and property.

Shujun Zhang received the Ph.D. degree from The State Key Laboratory of Crystal Materials, Shandong University, China, in 2000. He is currently Senior Research Associate at the Material Research Institute and Associate Professor in the Materials Science and Engineering Department of The Pennsylvania State University. He is an associate editor for the *Journal of the American Ceramic Society*. He holds one patent and has coauthored more than 200 papers in the area of functional single crystals and ceramics. He is now focusing on the structure–property–performance relationship of high-temperature, high-power, high-performance piezoelectric crystals and ceramics for sensor and transducer applications.

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Fig. 1. (solid line) Impedance and (dashed line) phase spectra of PZT samples as a function of normalized frequency depending on the width-to-thickness aspect ratio, G .

Fig. 2.
Temperature dependence of dielectric loss tangent of various polymer materials.

Fig. 3. Calculated electromechanical coupling factor and wave velocity of 1–3 piezocomposites with different polymer materials as a function of PZT volume fraction.

Fig. 4. Mechanical quality factor and electromechanical coupling of 1–3 PZT4/polymer (I) composites as a function of ceramic volume fraction: (solid line) modeled coupling factor of composite (I), (dashed lines) linear fitting of each parameter.

Fig. 5.

The change in resonant frequency and mechanical quality factor of 1–3 piezocomposites as a function of temperature.

TABLE I

Material Parameters for Various Polymer Materials.

Material	ρ (kg/m ³)	v_L (m/s)	v_s (m/s)	c_{11} (GPa)	c_{44} (GPa)	c_{12} (GPa)	ν^* (Poise)	σ	α (dB/m)	k (W/mK)	T_d (°C)
Polymer (I)	1135	2157	976	5.28	1.08	3.12	0.65	0.37	2360	0.3	—
Polymer (II)	1328	2237	880	6.46	1.0	4.46	5	0.41	1050	0.2	60
Polymer (A)	2164	2631	1514	14.98	4.96	5.06	14 to 22	0.25	4100	1 [*]	100
Polymer (B)	2196	3000	1588	19.76	5.54	8.68	150	0.31	5770	(1.2)4.5 [*]	60
Polymer (C)	2258	3130	1662	22.13	6.23	9.66	2000 to 2500	0.3	4740	1.12 [*]	—

Elastic properties and attenuations of the polymer materials were measured at 10 MHz for longitudinal and 5 MHz for shear properties. ν and T_d refer to the viscosity and thermal degradation temperature, respectively.

* Manufacturer's reported values.

TABLE II

Material Parameters of PZT4 Used in the Modeling.

ρ (kg/m ³)	e_{33} (C/m ²)	e_{31} (C/m ²)	$\epsilon_{33}^S / \epsilon_0 / \epsilon_0$	c_{11}^E	c_{12}^E	c_{13}^E	c_{33}^E
7700	13.84	-6.98	635	139	77.8	74.3	113

TABLE III

Measured Dielectric and Electromechanical Properties of Various 1–3 Piezocomposites.

Piezoelectric	Filler	d_{33} (pC/N)	$\tan \delta$	$e_{33}^S / e_0 / \epsilon_0$	k_t	v (m/s)	Q_m
PZT4	Polymer I	360	0.004	468	0.68	3952	200 (320*)
	Polymer II	350	0.005	487	0.68	3924	60 (150*)
	Polymer A	350	0.005	501	0.66	4019	30 (80*)
	Polymer B	360	0.004	480	0.63	3920	18 (55*)
	Polymer C	350	0.002	576	0.63	3850	25 (70*)
PZT8	Polymer I	220	0.003	546	0.6	4148	380 (440*)
	Polymer II	210	0.003	520	0.62	4064	100 (240*)
	Polymer A	200	0.004	502	0.6	4218	45 (50*)
	Polymer B	230	0.003	420	0.57	4305	13 (62*)
	Polymer C	220	0.006	440	0.55	3940	60 (50*)

Note that Q_m values outside parentheses were determined by equivalent circuit method using (7), whereas Q_m values in parentheses were determined from composite attenuation using (8).