# A study of the relationship between the rheo-dielectric effect and the elasticity of viscoelastic materials

Yiyan Peng and Lih-Sheng Turng<sup>a)</sup>

Department of Mechanical Engineering, University of Wisconsin–Madison, Madison, Wisconsin 53706

Haimei Li and Zhixiang Cui

School of Material Science and Engineering, Zhengzhou University, Henan 450002, China

(Received 28 December 2009; final revision received 30 November 2010; published 21 January 2011)

#### **Synopsis**

Dielectrostriction is a rheo-dielectric phenomenon that relates the variation of dielectric properties of a material to deformation. For an initially isotropic material, two independent material parameters, called the strain-dielectric coefficients,  $\alpha_1$  and  $\alpha_2$ , are required to describe dielectrostriction in terms of strain. Deformation affects a material's dielectric properties in two ways: (a) by introducing anisotropy in the material, which is characterized by  $\alpha_1$ , and (b) by changing the volume density of the polarizable species, which is associated with  $(\frac{1}{3}\alpha_1 + \alpha_2)$ . Purely viscous fluids remain isotropic during any flow-induced deformation, and therefore the coefficient  $\alpha_1$  is always zero. In this paper, the dielectrostriction effect is studied in viscoelastic materials that are formulated to possess different degrees of elasticity. A special planar capacitance sensor rosette was employed to measure the coefficients  $\alpha_1$  and  $\alpha_2$  for these viscoelastic materials. The relationship between the material's elasticity and the coefficient  $\alpha_1$  is discussed, together with some potential applications based on this relationship in the conclusion. © 2011 The Society of *Rheology*. [DOI: 10.1122/1.3539654]

# I. INTRODUCTION

Dielectrostriction, a rheo-dielectric phenomenon, describes the variation of dielectric properties of a material with deformation and is a fundamental property of any dielectric material [Stratton (1941); Landau *et al.* (1984); Shkel and Klingenberg (1998); Peng *et al.* (2005)]. Deformation affects the relative positions of the dipoles and the local electric field of the material, which leads to a change in dielectric properties.

Dielectric properties of a deformed isotropic material are described by a second order tensor,  $\varepsilon_{ij}$ , which can be approximated as a linear function of the strain tensor,  $u_{ij}$  [Stratton (1941); Landau *et al.* (1984)],

<sup>&</sup>lt;sup>a)</sup>Author to whom correspondence should be addressed; electronic mail: turng@engr.wisc.edu

<sup>© 2011</sup> by The Society of Rheology, Inc.

J. Rheol. 55(2), 301-311 March/April (2011)

$$\Delta \varepsilon_{ij} = (\varepsilon_{ij} - \varepsilon \,\delta_{ij}) = \alpha_1 u_{ij} + \alpha_2 u_{ll} \delta_{ij}, \tag{1}$$

where  $\alpha_1$  and  $\alpha_2$  are strain-dielectric coefficients, which are material parameters. Small variations of dielectric properties with deformation are required to justify the assumption of linearity in Eq. (1), namely,

$$\|\alpha_1 u_{ij}\| \ll \varepsilon, \quad |\alpha_2 u_{ll}| \ll \varepsilon, \tag{2}$$

where  $\varepsilon$  is the relative dielectric constant of the initially isotropic material. The double line brackets indicate the magnitude of the components in the tensor, while the single line brackets indicate the magnitude of the scalar.

The physical meaning of the strain-dielectric relationship [see Eq. (1)] can be clarified by introducing the strain deviator tensor,  $u_{ij}^{dev} = u_{ij} - \frac{1}{3}u_{ll}\delta_{ij}$ , which removes the volume effect from the first term in the right-hand side of Eq. (1), resulting in

$$\Delta \varepsilon_{ij} = \alpha_1 u_{ij}^{dev} + \left(\frac{1}{3}\alpha_1 + \alpha_2\right) u_{ll} \delta_{ij}.$$
(3)

One can see that deformations affect material's dielectric properties in two ways: (a) by introducing anisotropy in an initially isotropic material, which is characterized by  $\alpha_1$ , and (b) by changing the volume density of the polarizable species, which is associated with  $(\frac{1}{3}\alpha_1 + \alpha_2)$  [Shkel and Klingenberg (1998)]. Deformation-induced anisotropy can be observed in solids, but does not exist in viscous fluids. Because viscous fluids remain isotropic during any flow-induced deformation, the first strain-dielectric coefficient,  $\alpha_1$ , is always zero [Stratton (1941)]. The only dielectrostriction effect appearing in Newtonian liquids and gases is due to the variations of the dielectric properties with the density. Viscoelastic materials, such as polymers, show both viscous and elastic behaviors. Therefore, the observation of the dielectrostriction effect in viscoelastic materials can be expected.

By using a local field model, Shkel and Klingenberg (1998) derived strain-dielectric coefficients,  $\alpha_1$  and  $\alpha_2$ , for a mixture (or composite) with randomly distributed rigid inclusions under affine deformation,

$$\alpha_1 = -\frac{2}{5} \frac{(\varepsilon_{mix} - \varepsilon_c)^2}{\varepsilon_c},$$

$$\alpha_2 = -\frac{1}{3} \frac{(\varepsilon_{mix} - \varepsilon_c)(\varepsilon_{mix} + 2\varepsilon_c)}{\varepsilon_c} + \frac{2}{15} \frac{(\varepsilon_{mix} - \varepsilon_c)^2}{\varepsilon_c},$$
(4)

where  $\varepsilon_{mix}$  and  $\varepsilon_c$  are the relative dielectric constants of the mixture (or composite) and the continuum matrix medium, respectively. A pure elastic solid, which can be treated as a mixture of numerous polarizable molecules dispersed in a vacuum ( $\varepsilon_c$ =1) [Shkel and Klingenberg (1998)], is a common example of the above-mentioned material systems. Later, it was found experimentally that by taking  $\varepsilon_{mix} = \varepsilon$  (the relative dielectric constant of the polymer) and  $\varepsilon_c$ =1, these predictions also work well with purely solid polymers under small deformations [Lee *et al.* (2005)].

This study aims to observe the dielectrostriction effect in viscoelastic materials and explore the relationship between the dielectrostrictive response and the elasticity of the materials. A sensor rosette having two planar capacitance sensors was employed to measure the strain-dielectric coefficients of the materials. In the following sections, the concepts and background information for the dielectrostriction study utilizing the planar capacitance sensor are introduced. Then, the measurement of the strain-dielectric coeffi-



**FIG. 1.** (a) An initially isotropic material is subjected to a shear displacement  $u_y = \gamma z$ . Inter-digitated electrodes are located in the *xy*-plane and form an angle  $\theta$  with respect to the *y*-axis. (b) Each electrode strip has a width 2w and a length *l*, and is separated from each other by 2a.

cients,  $\alpha_1$  and  $\alpha_2$ , is presented. As an illustration, the first coefficient,  $\alpha_1$ , is compared for materials with varying degrees of elasticity during oscillatory shear flow experiments.

# **II. BACKGROUND**

 $\varepsilon_{effe}$ 

### A. Planar capacitance sensor for dielectrostriction

A planar capacitance sensor has been developed with inter-digitated electrodes deposited on a nonconductive substrate. When the planar capacitance sensor is placed adjacent to a dielectric material, a change in the dielectric properties of the material caused by the deformation could be characterized by the change in the capacitance of the sensor. A detailed examination of the rheological and electronic aspects of dielectrostriction measurements using planar capacitance sensors was discussed in [Peng *et al.* (2005); Peng (2008)]. The following gives a brief description of the concept of a planar capacitance sensor and a sensor rosette having two mutually perpendicular oriented sensors used to decouple and measure two strain-dielectric coefficients.

A planar capacitance sensor is formed by inter-digitated electrodes having equal width, W=2w, and being separated by a distance, A=2a, which is attached to a dielectric material, as shown in Fig. 1. Both the thickness of the dielectric material, h (which is not shown in Fig. 1), and the length of the electrode, l, are much larger than the electrode width and separation  $(h, l \ge W, A)$ . The electrodes in Fig. 1 are located in the *xy*-plane and form an angle,  $\theta$ , with respect to the *y*-axis. As derived by Peng (2008), the capacitance,  $C_{\theta}$ , of such a planar sensor is

$$C_{\theta} = \frac{1}{2} C_0 (\varepsilon_{effective} + \varepsilon_s),$$

$$C_{\theta} = [(\varepsilon_{xx} \varepsilon_{zz} - \varepsilon_{xz}^2) \cos^2 \theta + (\varepsilon_{yy} \varepsilon_{zz} - \varepsilon_{yz}^2) \sin^2 \theta + 2(\varepsilon_{xy} \varepsilon_{zz} - \varepsilon_{xz} \varepsilon_{yz}) \cos \theta \sin \theta]^{1/2},$$
(5)

where  $\varepsilon_s$  is the relative dielectric constant of the sensor substrate and  $\varepsilon_{effective}$  is the effective relative dielectric constant of the material on the top. For an isotropic material with relative dielectric constant,  $\varepsilon$ ,  $\varepsilon_{effective} = \varepsilon$ ; therefore, the capacitance of the sensor before deformation is  $C_{\theta} = \frac{1}{2}C_0(\varepsilon + \varepsilon_s)$ . The parameter *L* is the combined length of all electrode strips, and  $C_0$  represents the capacitance of the electrodes in free space. The

value of  $C_0$  can be obtained experimentally or estimated as  $C_0 = 2\varepsilon_0 L \ln(1+w/a)/\pi$ [Peng *et al.* (2005)], where  $\varepsilon_0$  is the permittivity of free space.

When the originally isotropic dielectric material is undergoing deformation, it becomes anisotropic, and its relative dielectric constant becomes a second rank symmetric tensor. According to the strain-dielectric relation in Eq. (1), the relative dielectric constant tensor can be expressed in terms of strain,

$$\boldsymbol{\varepsilon} = \begin{bmatrix} \varepsilon + \alpha_1 u_{xx} + \alpha_2 u_{ll} & \alpha_1 u_{xy} & \alpha_1 u_{xz} \\ \alpha_1 u_{xy} & \varepsilon + \alpha_1 u_{yy} + \alpha_2 u_{ll} & \alpha_1 u_{yz} \\ \alpha_1 u_{xz} & \alpha_1 u_{yz} & \varepsilon + \alpha_1 u_{zz} + \alpha_2 u_{ll} \end{bmatrix},$$
(6)

where  $u_{ll} = u_{xx} + u_{yy} + u_{zz}$ .

Substituting the components of the relative dielectric constant tensor into Eq. (5) yields the capacitance of the planar capacitor sensor under an arbitrary deformation,

$$C_{\theta} = \frac{1}{2} C_{0} \bigg( \varepsilon + \frac{\alpha_{1}(u_{xx}\cos^{2}\theta + u_{yy}\sin^{2}\theta + 2u_{xy}\sin\theta\cos\theta + u_{zz}) + 2\alpha_{2}u_{ll}}{2} \\ + \frac{\alpha_{1}^{2} [(u_{xx}\sigma_{zz} - u_{xz}^{2})\cos^{2}\theta + (u_{yy}u_{zz} - u_{yz}^{2})\sin^{2}\theta + 2(u_{xy}u_{zz} - u_{xz}u_{yz})\cos\theta\sin\theta]}{2\varepsilon} \\ + \frac{\alpha_{1}\alpha_{2}\sigma_{ll}(u_{xx}\cos^{2}\theta + u_{yy}\sin^{2}\theta + 2u_{xy}\cos\theta\sin\theta + u_{zz}) + \alpha_{2}^{2}u_{ll}^{2}}{2\varepsilon} \\ - \frac{(\alpha_{1}u_{xx}\cos^{2}\theta + \alpha_{1}u_{yy}\sin^{2}\theta + 2\alpha_{1}u_{xy}\sin\theta\cos\theta + \alpha_{1}u_{zz} + 2\alpha_{2}u_{ll})^{2}}{8\varepsilon} + \varepsilon_{s}\bigg). \quad (7)$$

Consider the dielectric material under a simple shear deformation,  $U_y = \gamma z$ , as shown in Fig. 1(a); the strain tensor can be expressed as

$$\mathbf{u} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & \gamma/2 \\ 0 & \gamma/2 & \gamma^2/2 \end{bmatrix}.$$
 (8)

The term  $\gamma^2/2$ , normally neglected in a simple shear flow, is sustained in this analysis since the overall dielectric effect of the deformation is quadratic with the shear intensity,  $\gamma$ . Substituting the above strain components into Eq. (7) and neglecting the higher-order of strain  $O(\gamma^4)$  yields the capacitance of a planar sensor under small shear deformation,

$$C_{\theta} = \frac{1}{2} C_0 \bigg( \varepsilon + \frac{2\varepsilon \alpha_1 + 4\varepsilon \alpha_2 - \alpha_1^2 \sin^2 \theta}{8\varepsilon} \gamma^2 + \varepsilon_s \bigg).$$
(9)

This expression shows that by conducting measurements with two differently oriented sensors (different values of  $\theta$ ) one can decouple strain-dielectric coefficients,  $\alpha_1$  and  $\alpha_2$ .

#### B. Sensor rosette for strain-dielectric coefficients

In this study, a sensor rosette, which consists of two identical planar capacitance sensors with mutually perpendicular oriented electrodes, was used to decouple and measure the strain-dielectric coefficients. The difference in capacitances of these two sensors expressed in terms of strain is



**FIG. 2.** (a) The rheometer produces oscillatory shear flow between concentric cylinders  $(\theta_0 = \gamma_0(R_2 - R_1)/R_1)$ . (b) Two planar sensors are attached to the fixed cylinder parallel-to-flow and perpendicular-to-flow. (c) The circuit detects the sensors' response. It is excited by 5 V AC voltage ( $V_0$ =7.07 V) at a frequency of 0.1 Hz. (d) The output signal is digitized, and its second harmonic signal is extracted from the fast Fourier transform. (e) The second harmonic signal is recorded by a perpendicular-to-flow sensor for silicone elastomer tested at a strain amplitude of 30%. Gaps in the data points are due to programmed stops of the rheometer head.

$$C_{\theta} - C_{\theta+\pi/2} = \frac{1}{2} C_0 \frac{\alpha_1^2 \cos 2\theta}{8\varepsilon} \gamma^2.$$
(10)

Equation (10) shows that the capacitance difference of these two sensors only involves the first strain-dielectric coefficient,  $\alpha_1$ . In addition, extracting signals from these two sensors is also helpful for temperature compensation and noise cancellation.

# **III. EXPERIMENT**

#### A. Material preparation

In this study, silicone oil, a purely viscous material, and silicone elastomer, a viscoelastic material, are mixed at different weight ratios to give varying degrees of elasticity. Silicone oil with a density of  $\rho$ =0.963 g/cm<sup>3</sup> (Aldrich Chemical) and liquid silicone elastomer with a density of  $\rho$ =1.08 g/cm<sup>3</sup> (GI 1110 from Silicone Inc.) were mixed at six different ratios, i.e., 10:0, 8:2, 6:4, 4:6, 2:8, and 0:10, by weight. Because silicone oil and silicone elastomer have similar chemical structures, they mix well with each other.

### B. Experimental setup

Figure 2 shows a schematic of the experimental setup of dielectrostriction measurements for the liquids. Figure 2(a) shows an AR-1000 rheometer from TA Instruments

**TABLE I.** The relative dielectric constant  $\varepsilon$  of silicone elastomer, silicone oil, and their mixtures at different ratios by weight and measured at 10 kHz.

Silicone elastomer: Silicone oil	10:0	8:2	6:4	4:6	2:8	0:10
ε	2.9	2.80	2.72	2.62	2.54	2.50

which provides an oscillatory shear flow between two concentric cylinders (Couette rheometer) and records the rheological responses. The external cylindrical container (with inner radius  $R_2=24$  mm) is fixed, and the inner cylinder (with radius  $R_1=22.5$  mm and immersed depth H=20.5 mm) is rotated by the rheometer. In Fig. 2(b), a sensor rosette is attached to the inner surface of the external cylinder with the electrodes of one sensor perpendicular to and the electrodes of the other sensor parallel to the flow direction. The width and distance between the two adjacent electrodes are 2w=2a=0.3 mm. The circuit in Fig. 2(c) is used to register small variations in the capacitance of the sensor [Doeblin (1990); Holman (1994); Horowitz and Hill (1998)] caused by the flow. The excitation voltage has an amplitude of 7.07 V alternating current (AC) voltage and a frequency of 0.1 Hz. A large resistance, R (~10 G $\Omega$ ), ensures that the charge, Q, on the sensor electrodes remains constant. By neglecting the effect of curvature of the sensor rosette mounted on the inner surface of the outer cylinder, the variation of voltage,  $\Delta V$ , across the electrodes is related to the change of capacitance,  $\Delta C$ , by

$$\frac{\Delta V}{V^0} = -\frac{\Delta C}{C} = -\frac{\Delta \varepsilon}{\varepsilon + \varepsilon_S},\tag{11}$$

where  $V^0$  is the voltage between the electrodes before deformation and  $\Delta \varepsilon$  is the variation of dielectric constant with deformation.

In an ideal case,  $\varepsilon_s$  represents the relative dielectric constant of the electrode substrate. In reality,  $\varepsilon_s$  accounts for the additional capacitance due to other fixtures. In this study the effective relative dielectric constant of the substrate was estimated as  $\varepsilon_s$ =47.73. This value accounts for the fringe field coupling through the thin polyimide substrate of the sensor electrodes and the aluminum wall of the external cylinder. The relative dielectric constant,  $\varepsilon$ , of each sample measured at 10 kHz is tabulated in Table I. A detailed description of the dielectric measurements and the estimation of the effective dielectric constants are provided in [Peng *et al.* (2006)].

The voltage outputs due to the dielectrostriction response measured by the electrodes parallel to the flow direction,  $\theta = 0^{\circ}$ , and perpendicular to the flow direction,  $\theta = 90^{\circ}$ , in terms of strain are

$$\Delta V_{\parallel} = -\frac{2\alpha_1 + 4\alpha_2}{8(\varepsilon + \varepsilon_s)}\gamma^2 V^0, \quad \Delta V_{\perp} = -\frac{2\varepsilon\alpha_1 + 4\varepsilon\alpha_2 - \alpha_1^2}{8\varepsilon(\varepsilon + \varepsilon_s)}\gamma^2 V^0.$$
(12)

These equations show that the variations of voltage due to a shear flow are quadratic with the shear strain,  $\gamma = \gamma_0 \sin(2\pi f)t$ . Therefore, the dielectrostriction response should be measured at the double frequency of the mechanical oscillations, 2f. To compare the dielectrostriction responses in different systems, all testing results will be presented in the same form, where only the material properties are retained,



**FIG. 3.** Measured strain-dielectric relationships of pure silicone elastomer observed by the (a) perpendicularto-flow and (b) parallel-to-flow sensors. The experimental error for each data point is estimated within 10%.

$$\frac{\Delta \varepsilon_{\parallel,\perp}}{\varepsilon} = \left(\frac{\varepsilon_{eff(\parallel,\perp)} - \varepsilon}{\varepsilon}\right) = -\frac{\Delta V_{\parallel,\perp}(\varepsilon + \varepsilon_s)}{V^0 \varepsilon}.$$
(13)

Extracting dielectrostriction signals from the two mutually perpendicular sensors yields the first strain-dielectric coefficient,

$$|\alpha_1| = \frac{1}{\gamma} \sqrt{8\varepsilon(\varepsilon + \varepsilon_s)(\Delta V_\perp - \Delta V_\parallel)/V^0}.$$
(14)

The second strain-dielectric coefficient,  $\alpha_2$ , can be obtained by plugging  $\alpha_1$  into either of the expressions in Eq. (12).

# C. Test procedure

All samples were tested under the time sweep mode and at a mechanical oscillation frequency of 8 Hz. The dielectrostriction response of the pure silicone elastomer was measured at the strain amplitude,  $\gamma_0$ , which ranged from 10% to 30% with an increment of 5%. For pure silicone oil and the mixtures of silicone elastomer and silicone oil, the dielectrostriction response was measured at a strain amplitude of 30%. Voltage variations,  $\Delta V_{\perp}$  and  $\Delta V_{\parallel}$ , were digitally recorded at a sampling rate of 100 000 Hz. The output signal was obtained through a fast Fourier transform of the measured data. The strain and the corresponding phase shift,  $\delta$ , were recorded by the rheometer simultaneously with the voltage outputs. Each material mixture was loaded into the rheometer twice and tested at least three times.

# **D. Experimental results**

The voltage outputs,  $\Delta V_{\perp}$  and  $\Delta V_{\parallel}$ , were obtained using the procedure outlined in Fig. 2 and described in Sec. III B. Figure 2(e) gives sample sets of the data: silicone elastomer tested at 30% strain amplitude, and the second harmonic signal recorded by the perpendicular-to-flow sensor.

Figure 3 shows the strain-dielectric relationship of pure silicone elastomer at a strain amplitude ranging from 10% to 30%. The strain-dielectric relationship of pure silicone elastomer and the mixtures of silicone elastomer and silicone oil at different weight ratios at a strain amplitude of 30% are shown in Fig. 4. The ratios of  $\Delta \varepsilon_{\perp} / \varepsilon \equiv (\varepsilon_{eff}(\pi/2) - \varepsilon)/\varepsilon$  [cf. Figs. 3(a) and 4(a)] and  $\Delta \varepsilon_{\parallel} / \varepsilon \equiv (\varepsilon_{eff}(0) - \varepsilon)/\varepsilon$  [cf. Figs. 3(b) and 4(b)] express the variations of the relative dielectric constants, obtained through the



**FIG. 4.** Measured strain-dielectric relationships of pure silicone elastomer and mixtures of silicone oil and silicone elastomer at various weight ratios obtained by (a) perpendicular-to-flow and (b) parallel-to-flow sensors. The experimental error for each data point is estimated within 10%.

perpendicular-to-flow sensor and the parallel-to-sensor, respectively, divided by the relative dielectric constant of the material before deformation. These ratios are deduced from Eq. (13) and presented in terms of the square of strain amplitude,  $\gamma_0^2$ . The measurement for pure silicone oil was also conducted at a mechanical oscillation frequency of 8 Hz and a strain amplitude of 30%. However, silicone oil demonstrates no measurable dielectrostriction response. This is in agreement with the expectation since silicone oil is a purely viscous material; hence the dielectrostriction response of silicone oil can only be due to the change of density, which might be too weak to be observed with the present experimental setup.

The phase shift,  $\delta$ , of the pure silicone elastomer at a strain amplitude from 10% to 30% is shown in Fig. 5(a). Figure 5(b) gives the phase shift,  $\delta$ , of pure silicone elastomer and the mixtures of silicone oil and silicone elastomer at various weight ratios at a strain amplitude of 30%. Note that when the strain increases, the phase shift increases; that is, the elasticity of the material decreases [cf. Fig. 5(a)]. In addition, the elasticity of the material decreases in the weight fraction of the silicone oil [cf. Fig. 5(b)].



**FIG. 5.** Measured phase shift  $\delta$  of (a) silicone elastomer at various strain amplitudes and (b) silicone elastomer, and the mixtures of silicone oil and silicone elastomer at various weight ratios at a strain amplitude of 30%.



**FIG. 6.** The first strain-dielectric coefficient,  $\alpha_1$ , and the combination coefficient  $(\frac{1}{3}\alpha_1 + \alpha_2)$ , [(a) and (b)] for pure silicone elastomer with varying strain amplitudes and [(c) and (d)] for pure silicone elastomer, and the mixtures of silicone oil and silicone elastomer, at a strain amplitude of 30%.

# **IV. DISCUSSION**

It should be pointed out that the experimental setup used in this study does not provide the sign of the strain-dielectric coefficients. However, the sign of these two coefficients can be determined using the following approach. It is known that compression would increase the number density of polarizable inclusions in the material, thus increasing its dielectric constant. The reverse is true for inclusions having lower dielectric constants than the matrix material. Because compressive strains have a negative sign, only a negative value of  $(\frac{1}{3}\alpha_1 + \alpha_2)$  will be meaningful according to Eq. (3). Thus, in this study, first both positive and negative  $\alpha_1$ 's have been assumed for all samples by extracting signals from the perpendicular-to-flow and parallel-to-flow sensors [cf. Eq. (14)]. Then the coefficient,  $\alpha_2$ , was calculated for each value of  $\alpha_1$  having either a positive or a negative sign. Only the value of  $\alpha_1$  that produces a negative combination  $(\frac{1}{3}\alpha_1 + \alpha_2)$  is considered to be valid. It was found that a positive  $\alpha_1$  produced a negative combination  $(\frac{1}{3}\alpha_1 + \alpha_2)$ . Figure 6 presents the first strain-dielectric coefficient,  $\alpha_1$  [cf. Figs. 6(a) and 6(c)], and the combination coefficient,  $(\frac{1}{3}\alpha_1 + \alpha_2)$  [cf. Figs. 6(b) and 6(d)], for pure silicone elastomer with varying strain amplitudes, and pure silicone elastomer and mixtures of silicone oil and silicone elastomer at a strain amplitude of 30%, respectively. It is interesting to see

**TABLE II.** Strain-dielectric coefficient  $|\alpha_1^{(e)}|$  of elastic solid counterparts of silicone oil, silicone elastomer, and their mixtures at various weight ratios, as predicted by Eq. (4).

Silicone elastomer: Silicone oil	10:0	8:2	6:4	4:6	2:8	0:10
$ \alpha_1^{(e)} $	1.44	1.29	1.19	1.06	0.95	0.90

that while  $\alpha_1$  is predicted to be a negative value for elastic solids [see Eq. (4)], it turned out to be positive for viscoelastic liquids according to experimental measurements. The factors that change the sign of  $\alpha_1$  are not clear at this stage and require further study.

According to Eq. (4),  $\alpha_1$  is strongly related to the dielectric constant of each material system. To compensate for the dielectric constant difference among different materials and single out the influence of the degree of elasticity, Eq. (4) is employed to predict the strain-dielectric coefficient,  $\alpha_1^{(e)}$ , of the elastic solid counterpart of each liquid material system with the same relative dielectric constant,  $\varepsilon$ , as listed in Table I. By taking  $\varepsilon_{mix} = \varepsilon$  and  $\varepsilon_c = 1$ ,  $|\alpha_1^{(e)}|$  is calculated and listed in Table II. Then the measured strain-dielectric coefficient of liquid material systems was divided by the predicted strain-dielectric coefficient of their elastic solid counterparts, whose absolute value,  $|\alpha_1/\alpha_1^{(e)}|$ , is presented in Fig. 7. The corresponding value,  $\cos \delta (=G'/G^*)$ , which reflects the significance of the elastic component [Bird *et al.* (1987)], is also shown in Fig. 7 for comparison. Note that G' and  $G^*$  are the storage and complex moduli, respectively.

From Fig. 7, one can see that (1) for pure silicone elastomer,  $|\alpha_1/\alpha_1^{(e)}|$  decreases with an increase in strain amplitude; and (2) the value of  $|\alpha_1/\alpha_1^{(e)}|$  gradually decreases with an increase of the weight fraction of the viscous material—silicone oil. These two findings are attributed to the fact that the viscous component in the material remains isotropic during any flow-induced deformation, thus not contributing to  $\alpha_1$ . Moreover, it is interesting to see that  $|\alpha_1/\alpha_1^{(e)}|$  has a similar trend as  $\cos \delta (=G'/G^*)$  for both silicone elastomer with varying strain amplitudes [cf. Fig. 7(a)] and for mixtures with varying weight fractions of silicone oil [cf. Fig. 7(b)]. This trend suggests that  $\alpha_1/\alpha_1^{(e)}$  might be used as an alternative parameter to estimate the elasticity of the material or its variation due to, say, phase change, curing, or any other cause. However, how to quantitatively relate the parameter,  $\alpha_1/\alpha_1^{(e)}$ , to the elasticity, such as with phase shift,  $\delta$ , is not fully understood yet and needs to be explored in further studies.



**FIG. 7.** The absolute value of the measured strain-dielectric coefficient of liquid material systems was divided by the predicted strain-dielectric coefficient of their elastic solid counterparts,  $|\alpha_1/\alpha_1^{(e)}|$ , and  $\cos \delta$  for (a) silicone elastomer at various strain amplitudes and (b) silicone elastomer, and the mixtures of silicone oil and silicone elastomer, at a strain amplitude of 30%.

Dielectrostriction describes the change in dielectric properties with deformation. For an originally isotropic material, two strain-dielectric coefficients,  $\alpha_1$  and  $\alpha_2$ , are required to relate the dielectric responses with strain. The first strain-dielectric coefficient,  $\alpha_1$ , is responsible for the change in dielectric properties via deformation-induced anisotropy. This paper presents a preliminary study on the elasticity dependence of the straindielectric coefficient,  $\alpha_1$ , in various viscoelastic materials and the potential application of measuring the material's elasticity through the dielectrostriction effect. It has been found that the ratio of the strain-dielectric coefficient of the viscoelastic material to the straindielectric coefficient of its elastic solid counterpart,  $\alpha_1/\alpha_1^{(e)}$ , varies with the material's elasticity. This indicates that  $\alpha_1/\alpha_1^{(e)}$  might be used as an alternative parameter to estimate the elasticity of the material or its variation and to provide a novel approach to study the viscoelastic properties of materials.

# ACKNOWLEDGMENTS

The authors would like to thank Professor Yuri M. Shkel at the University of Wisconsin–Madison for initial guidance and Yuxi Cheng at the University of Wisconsin–Madison for help with dielectric constant measurement. This research was supported by the Graduate School of the University of Wisconsin–Madison.

#### References

- Bird, R. B., C. F. Curtiss, R. C. Armstrong, and O. Hassager, *Dynamics of Polymeric Liquids: Kinetic Theory* (Wiley-Interscience, New York, 1987), Vol. 2.
- Doeblin, E. O., Measurement Systems: Application and Design (McGraw-Hill, New York, 1990).
- Holman, J. P., Experimental Methods for Engineers (McGraw-Hill, New York, 1994).
- Horowitz, P., and W. Hill, The Art of Electronics (Cambridge University Press, London, 1998).
- Landau, L. D., E. M. Lifshitz, and L. P. Pitaevskii, *Electrodynamics of Continuous Media* (Pergamon, New York, 1984).
- Lee, H. Y., Y. Y. Peng, and Y. M. Shkel, "Strain-dielectric response of dielectrics as foundation for electrostriction stresses," J. Appl. Phys. 98, 074104 (2005).
- Peng, Y. Y., "Rheo-dielectric studies in polymeric systems," Ph.D. dissertation, University of Wisconsin– Madison, 2008.
- Peng, Y. Y., D. J. Prochniak, and Y. M. Shkel, "Rheo-dielectric study in polymeric nano-suspensions," Proceedings of IMECE 2006, 449–456 (2006).
- Peng, Y. Y., Y. M. Shkel, and G. H. Kim, "Stress dielectric response in liquid polymers," J. Rheol. 49, 297–311 (2005).
- Shkel, Y. M., and D. J. Klingenberg, "Electrostriction of polarizable materials: Comparison of models with experimental data," J. Appl. Phys. 83, 7834–7843 (1998).
- Stratton, J. A., Electromagnetic Theory (McGraw-Hill, New York, 1941).