A comment on "Observation of the inverse piezoelectric effect in polyethylene while under a polarizing field" by R. W. Greaves and D. R. Lamb

Greaves and Lamb (GL) [1] reported that they have observed the converse (or inverse) piezoelectric effect in polyethylene. This conclusion is based upon the authors' experimental data which they believe exclude the electrostrictive effect, because the observed behaviour for polyethylene is linear and dependent upon the direction of the applied polarizing field. We suggest here, however, that GL's results probably should be interpreted as arising from the electrostrictive effect in polyethylene.

The converse piezoelectric effect can be defined in terms of the piezoelectric strain coefficient  $d_{ii}$  as

$$\epsilon_j = d_{ij} E_i, \tag{1}$$

where  $E_i$  is the applied electric field, and  $\epsilon_j$  is the induced strain. Equation 1 describes the linear (first order) piezoelectric effect, for which the strain is proportional to the electric field. When the electric field is large, however, quadratic terms must be considered in addition to the linear terms, and in this case, Equation 1 becomes

$$\epsilon_j = d_{ij}{}^0 E_i + \gamma_{ijk} E_i E_k, \qquad (2)$$

where  $d_{ij}^{0}$  is the value of  $d_{ij}$  for infinitesimal fields. The first term describes the converse piezoelectric effect, whereas the second term represents the electrostrictive effect;  $\gamma_{ijk}$  is the electrostrictive coefficient. For their study of polyethylene, GL defined the 3-direction as the axis of polarization. In this co-ordinate system, Equation 2 can be simplified:

$$\epsilon_3 = (d_{33}^0 + \gamma E_3) E_3, \tag{3}$$

where  $\gamma = \gamma_{333}$ .

In GL's measurements, an electric field given by

$$E_3 = E_{\rm dc} + E_{\rm ac} \tag{4}$$

was applied to the polymer;  $E_{dc}$  representing the large dc polarizing field and  $E_{ac}$  a small alternating field. Substituting Equation 4 into Equation 3 and neglecting products of small quantities, we can write:

$$\epsilon_3 \cong (d_{33}^0 + \gamma E_{dc}) E_{dc} + (d_{33}^0 + 2\gamma E_{dc}) E_{ac}.(5)$$

By monitoring the rms signal voltage  $V_2$  from a PZT ceramic receiver, GL detected only the ac contributions to the total strain  $\epsilon_3$ . In terms © 1973 Chapman and Hall Ltd. of these ac components, Equation 5 becomes

$$\varepsilon_3 (\mathrm{ac}) \cong (d_{33}^0 + 2 \gamma E_{\mathrm{dc}}) E_{\mathrm{ac}}. \tag{6}$$

From the analysis of GL,

$$\epsilon_3 (ac) = k V_2 t^{-1},$$
 (7)

where t is the thickness of the polyethylene sample, and for the experimental conditions employed by GL,

$$k = 1.88 \times 10^{-11} \,\mathrm{C}\,\mathrm{N}^{-1}$$
. (8)

Finally, by combining Equations 6, 7 and 8, we can write

$$(d_{33}^{0} + 2 \gamma E_{dc}) \cong (1.88 \times 10^{-11}) (V_2/V_1),$$
 (9)

because  $V_1 = E_{ac} t$ .

It is evident from Equation 9 that the signal associated with the electrostrictive effect depends upon the direction of the applied polarizing field; moreover, the electrostrictive effect is consistent with the linear dependence of  $V_1$  versus  $V_2$  depicted by GL in Fig. 5.

The fact that the data points portrayed by GL in Fig. 4 do not pass through the origin has been attributed by them to noise in their receiving equipment, but the correct explanation of this behaviour may be more complex. For  $E_{de}$  > 50 kV cm<sup>-1</sup>, however, the most obvious interpretation is that the data depicted in Figs. 4 and 6 represent the electrostrictive effect in polyethylene. In light of these arguments, we have used the data of GL to determine the electrostrictive coefficient  $\gamma$  for polyethylene. The value that we have obtained ( $\gamma = 1.7 \times 10^{-20} \text{ m}^2 \text{ V}^{-2}$ ) is comparable in magnitude with that observed for another organic polymer, polyvinylidene fluoride films ( $\gamma = 3.9 \times 10^{-20} \text{ m}^2 \text{ V}^{-2}$ ) [2]. Thus, we conclude that the electrostrictive effect in polyethylene probably accounts for the observations reported by GL as the inverse piezoelectric effect.

## References

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- 2. HIEJI KAWAI, Oyo Butsuri 39 (1970) 413.

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