BRIEF COMMUNICATION

MEASUREMENT OF THE PYROELECTRIC COEFFICIENT AND PERMITTIVITY ON RHODODENDRON AND ENCEPHALARTOS LEAVES AND ON THE INSECT *PERIPLANETA AMERICANA*

M. Simhony, *The Racah Institute of Physics, The Hebrew University, Jerusalem 91000 Israel, and Center for Laser Studies, University of Southern California, Los Angeles, California 90007 U.S.A.*
H. Athenstaedt, *Institut für Molekular-Physikalische Physiologie, Hindenburgstrasse 2-4, D-3000 Hannover, West Germany*

ABSTRACT Pyroelectric (PE) voltage responses to single square radiation signals absorbed in epidermal layers of the leaves of *Encephalartos villosus* and *Rhododendron catawbiense* and in the integument of the insect *Periplaneta americana L.* were investigated on dry preparations as well as on whole fresh leaves and live insects. Measurements of the initial slopes of the responses, the rise times, and the peak voltages were carried out with 10 interchangeable load resistors from 5MΩ to 10 GΩ, connected parallel to the samples. In encephalartos leaves this yielded for the permittivity ϵ and the PE coefficient λ in the upper/lower epidermis, $\epsilon = 30/60$ and $\lambda = 2/6$ pC/cm^{2}K, respectively, and in rhododendron, $\epsilon = 60/200$ and $\lambda = 3/15$ pC/cm^{2}K. In periplaneta we found $\epsilon = 300$ and $\lambda = 20$ pC/cm^{2}K for the abdomen rings, and $\epsilon = 1{,}000$ and $\lambda = 350$ pC/cm^{2}K for the thorax of live insects. Generally, dry preparations of leaves, and especially of insects, showed smaller PE responses. The outer epidermis of live plant leaves and the integument of live insects appear to be polar and to exhibit pronounced PE properties.

Investigation of the pyroelectric (PE) effect (1) in various animal and plant tissues was pioneered by Lang (2) and Athenstaedt (3–7) and additional studies have been made by Liboff and Furst (8, 9). An in-depth quantitative investigation of the PE effect in leaves of the palm-like plant *Encephalartos villosus* was recently carried out by Lang and Athenstaedt (10) using the analysis by Simhony and Shaulov of the PE responses to step (11) and square (12) radiation signals.

The assertion that the PE effect plays an important role in living organisms is based on distinctive features, such as its dependence on dT/dt, rather than on ΔT or grad T, which results in a zero PE voltage when $dT/dt = 0$, independent of how far from ambient the sample temperature T might be. This feature could have served to distinguish the PE effect, but, has become a source of confusion, instead, due to the use of improperly shaped and modulated heating or radiation signals. Because of the differentiation with time, the PE response is then a complex mixture of sample and signal characteristics; it might be just as hard to prove the PE character of a response as it is to ascribe the response to any other voltage producing phenomenon.

Faced with these difficulties, Simhony and Shaulov have characterized parameters of PE voltage responses to radiation signals of different shapes, durations, and repetition rates as functions of the parameters of both the signals and the PE samples. Their analysis for step (11) and square single (12) radiation patterns provides a method to determine the PE character of a response.

We have applied this method to study the PE effect in live insects and in wet leaves, using a new technique of nondestructive electroding.[1] The results show for the first time that the PE effect exists in living organisms. In most cases the PE voltage responses of live samples were higher than those of dry preparations, indicating that the PE effect plays a role in living organisms.

From the obtained PE voltage responses in both the live samples and the dry preparations, we derive the permittivity ϵ and the PE coefficient λ of the polar layers in which the PE effect originated. For this we use a previously described method (13) and independently measured values of the volume specific heat c. The values of ϵ and λ so obtained are reasonable as well as important in that this is the first time that such estimates are reported for live samples.

THEORETICAL BACKGROUND

In an unclamped sample having spontaneous polarization P_s, in which the flux F from a step radiation signal is absorbed uniformly, causing a small temperature change ΔT, ϵ, λ, and c can be considered constant, and ϵ is given by (13)

$$\epsilon = C_s L/\epsilon_0 A = \tau_e L/\epsilon_0 A R_L. \tag{1}$$

Here L, A, and C_s are, respectively, the thickness, the front electrode area, and the electrical capacitance of the sample, and $\epsilon_0 = 8.85$ pF/m; R_L is the load resistance, connected parallel to the sample, and τ_e is the electronic RC time constant of the circuit (Fig. 1). R_L and the input capacitance C_i of the preamplifier are sufficiently smaller than the resistance R_s and capacitance C_s of the sample, so that $\tau_e = R_L C_s$. Also, τ_e should be sufficiently smaller than the thermal time-constant τ_T of the sample. Then τ_e can be obtained from the PE response $V(t)$ as its rise-time t_r, i.e., the time needed to reach $1 - e^{-1} = 63\%$ of the peak voltage V_p (see Fig. 2, curve $V'(t)$ for $\tau_T = 500\ \tau_e$). Otherwise, (Fig. 2, response $V(t)$ for $\tau_T = 5\ \tau_e$) t_r is shorter than τ_e, and can be obtained from Fig. 3 of reference 13. The PE coefficient λ can be obtained from the initial slope k of the PE response (Fig. 2),

$$\lambda = \epsilon_0 \epsilon c k/F, \tag{2}$$

with $\lambda \equiv |\mathrm{d}P_s/\mathrm{d}T|$, and $k \equiv [\mathrm{d}V(t)/\mathrm{d}t]_{t=0}$. Also, because $k = HV_p/\tau_e$, where $H \equiv (\tau_e/\tau_T)$ $\exp[\tau_t/(\tau_e - \tau_T)]$, one can obtain λ from the peak PE voltages V_p, using

$$\lambda = cLHV_p/AR_L F. \tag{3}$$

The fall time of $V(t)$ yields τ_T, if the step irradiation is longer than at least $3\tau_T$. If such a

---

[1] Simhony, M., and H. Athenstaedt. Unpublished work.

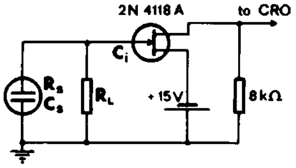

FIGURE 1 Measuring circuit.

lengthy irradiation causes too much heating, so that ϵ, λ, and c cannot be considered constant, or if the prolonged heating results in appreciable non-PE currents, then a single square pulse of duration $\mathcal{T}$ longer than the peaking time t_p, $t_p < \mathcal{T} < \tau_T$, can be used instead (see Fig. 2). The switching off of the radiation signal at $t = \mathcal{T}$ is a step dark signal. The PE response to this (marked "square" on Fig. 2) yields the same values of $|k|$, τ_e, and $|V_p|$ (measured down from the step $V(t)$ response), provided that $\Delta T(\mathcal{T})$ is small. The time constant (fall-time t_f) of the slow return of this response to the zero voltage line yields τ_T (12).

EXPERIMENTAL TECHNIQUES

The samples were of four types: (*a*) Small (~1 cm^2) pieces of upper or lower epidermis, prepared by scraping away the opposite epidermal layer and the soft and wet internal (ionically conductive) tissue. An electrode (10 or 25 mm^2) was then applied to the inner and outer surface. These samples dried out during preparation and so were considered "dead." (*b*) Large (~5 cm^2) parts of freshly cut whole

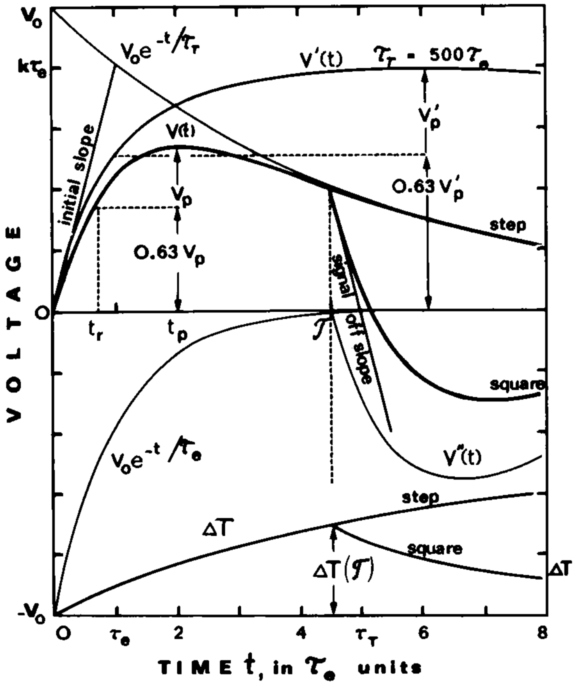

FIGURE 2 Theoretical PE responses. ΔT is temperature change of sample due to absorbed step or square radiation signal. PE response $V(t)$ is sum of $V_0 \exp(-t/\tau_T)$ and $-V_0 \exp(-t/\tau_e)$. After end of square pulse, PE response is sum of $V(t)$ and the negative response $V''(t)$ to the step dark signal at $t = \mathcal{T}$.

leaves having both the dark and the radiation receiving electrodes (25 or 50 mm^2) on one, upper or lower, epidermis. The measurements could be finished within 30 min after cutting the leaf off the plant. These samples are in that sense considered "in vivo." (*c*) Small (~5 × 10 mm) pieces from the abdominal rings of the insect, prepared by scraping away the muscle tissue from the inner side, then applying an electrode to the outer and inner sides (6). (*d*) In vivo samples on the thorax of live insects. The insects were attached with adhesive tape to the sample holder so that they could breathe and move their legs, head, and abdomen, but not the thorax. The two electrodes (10 mm^2 each) were then applied to the same surface of the thorax. Preparation and measurement took ~1 h, after which the insects showed no ill effects.

Electrodes were mostly water-suspended graphite, held in place with adhesive rings. Connection to the contacting poles of the teflon sample holder was made with aluminium foil leads and silver paste. Other electrodes were also tried but yielded a lower response (footnote 1). The sample holders were mounted in a cryostat with a quartz window allowing the radiation from a high pressure xenon or quartz-halogen lamp to be focused on the whole electrode of the sample by a quartz lens. The impinging radiation flux was measured by replacing the sample with a photometer head (40× optometer; United Detector Technology Inc., Santa Monica, Calif.). The flux absorbed with the water-suspended graphite electrode was approximated as 90% of the impinging flux. The step and square radiation signals were obtained with an electronically controlled shutter (Prontor), whose opening and closing times with a diaphragm diameter of 1.3 cm was 0.3 and 0.2 ms. This was, as required for a step signal, at least 10 times shorter than τ_e; τ_e was controlled by a set of 10 interchangeable resistors R_L from 5 MΩ to 10 GΩ. The signal from the preamplifier (Fig. 1) was observed (and photographed) on the screen of a Tektronix 5103N cathode ray oscilloscope with 5A22N vertical differential amplifier and 5B12N time-base plug-in's (Tektronix, Inc., Beaverton, Oreg.). PE responses down to 10 µV could be measured on the 20-µV/div sensitivity setting (~0.5 div peak-to-peak noise).

EXPERIMENTAL RESULTS

Fig. 3 shows six types (*a–f*) of PE response traces obtained in the samples. Type *a* (encephalartos #8; whole, upper epidermis) serves mostly reconnaissance purposes. The almost perfectly symmetric reversal of the response to "signal off" indicates the overwhelmingly PE

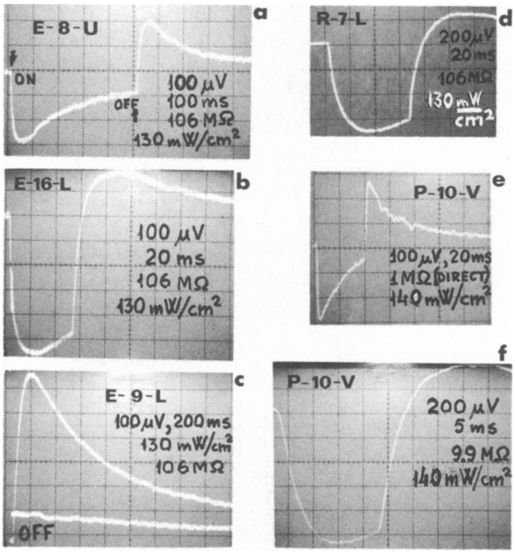

FIGURE 3 Survey of PE response traces.

character of the response (12) during up to 600 ms (usually it is not so long). The fall times of the "signal on" and "off" responses yield τ_T ~300 ms. From responses similar to type *b* (encephalartos #16 whole; "live," lower epidermis) one obtains a more accurate measure of V_p and the 0.63 V_p, needed for the τ_e measurement. On pictures of type *c* the "signal on" response is below the screen, which is filled by the "signal off" response for an accurate measure of τ_T (the lower line is the prolongation of the response; this long decay is a non-PE component). Response of type *d* (rhododendron #7; whole, lower epidermis) and *f* (periplaneta #10; in vivo) are useful in estimating t_r and τ_e (e.g., ~8 ms in rhododendron #7 and ~5 ms in periplaneta #10); for a measurement, the horizontal scale was made four times faster. To measure *k* we use the most sensitive *Y* scale with minimum possible noise distortion, and increase the horizontal speed until the initial response is seen to rise by an angle of ~45° (for maximum *X* and *Y* axis accuracy). Fig 3 *e* shows that in the live insect the PE response is so large that it can be observed by a direct hook-up to the oscilloscope.

The results of our measurements are presented in Table I. The thickness *L* of the polar (PE) layer was assumed to be the thickness of the corresponding epidermal (or integument) sample obtained after carefully scraping away the opposite layer and the soft wet inner tissues exhibiting ionic conductivity. In insects it was shown by carefully grinding away thin layers, layer after layer, off the integument, that the polarization is uniform throughout the whole thickness (6). Therefore, the *L* values are valid. In the plant epidermis however such an experiment was not carried out. (In "in vivo" samples having both electrodes on one side the layer thickness is entered twice [12]).

The *c* values were obtained with a Perkin-Elmer DSC differential calorimeter (Perkin-

TABLE I
EXPERIMENTAL RESULTS

	Layer Thickness (*L*)	Specific heat (*c*)	Permittivity (ϵ)	PE coefficient (λ)	PE figure of merit	
					k/*F*	($\lambda/\epsilon_0\epsilon c$)
	(μm)	(j/cm^3K)		(pC/cm^2K)	$\left(\frac{mV/s}{W/cm^2}\right)$	(10^{-3} cm^2/C)
Encephalartos leaves						
Upper epidermis: whole	—	1.9	30	2	300	400
scraped layer	78	1.4	3.5	0.5	1,000	1,200
Lower epidermis: whole	—	1.9	60	6	400	600
scraped layer	74	1.4	100	3	200	250
Rhododendron leaves						
Upper epidermis: whole	—	2.0	60	3	200	280
scraped layer	33	1.4	300	2	30	40
Lower epidermis: whole	—	2.0	200	15	400	420
scraped layer	60	1.4	*	*	*	*
Periplaneta						
Abdomen rings: scraped	70	1.3	300	20	600	580
Thorax: scraped	90	1.6				
In vivo	—	—	1,000	350	2,100	2,500
Triglycine sulphate (TGS)	—	1.7	30	$3 \cdot 10^4$	—	$5 \cdot 10^6$
Strontium (0.7)-barium (0.3) niobate (SBN)	—	2.5	1,700	10^5	—	$3 \cdot 10^5$

*Always cracked; not measurable.

Elmer Corp., Norwalk, Conn.) on small (a few milligrams) pieces of the material. It was assumed that the c values of the whole leaves are identical with those of the polar layers. The values of ϵ and λ are averages obtained on several samples of each material. The deviations from these values did not exceed 30% in individual samples, which should be considered good for biological materials. The PE figure of merit (11, 14) of the materials was calculated twice: as the speed of the initial rise k of the PE response per unit absorbed radiation flux, and also from the obtained values of the material parameters ϵ, λ, and c. The two values show remarkable agreement, considering the different measurables from which they were obtained, and the many assumptions made in the theory, which certainly do not always hold. For comparison, Table I also shows data for triglycine sulphate (TGS) single crystals, a material with the highest known figure of merit, and for strontium-barium niobate (SBN).

CONCLUSIONS

We have obtained voltage responses to step and square radiation signals absorbed in epidermal layers of the leaves of *Encephalartos villosus* and *Rhododendron catawbiense* and in the integument of the insect *Periplaneta americana L.* We have shown that these responses, though complex, remain overwhelmingly pyroelectric during quite long periods of time, at least 10 ms and up to ~400 ms, depending on material, sample preparation, and conditions of measurement. The PE character of the responses was shown, among others, by the symmetric reversibility to on and off signals, by the identity of the slopes at the rise and at the end of the signal, and by their independence of R_L in the whole range from 5 MΩ to 10 GΩ. Therefore the measurements allowed to derive sensible values of the dielectric constant and the PE parameters. We have developed a technique allowing measurements on live insects and whole fresh leaves. The live samples exhibit larger PE responses than the dry preparations. We conclude that the outer epidermal layers of live plant leaves and insects, at least of these investigated, are polar and exhibit pyroelectric properties.

This work was carried out while Dr. Simhony was a guest scientist at the Institut für Molekular-Physikalische Physiologie (IMP) in Kiel, on a stipend and travel grant from the Deutscher Akademischer Austauschdienst (DAAD), Bonn, West Germany. The time-consuming revision of the manuscript was performed while working under a grant from the U.S. Army Research Office, Durham, N.C., for the study of pyroelectricity in thin samples at the University of Southern California Center for Laser Studies, Los Angeles.

Mr. B. Raabe, electrical engineer, and Ms. C. Gerbrecht and Ms. C. Mehrens, medical technicians of IMP provided excellent help in circuit fixing, adjustment of equipment, sample preparation, measurements, and calculations. We extend our appreciation and thanks to all.

Received for publication 15 May 1979 and in revised form 24 September 1979.

REFERENCES

1. Lang, S. B. 1974. Sourcebook of Pyroelectricity. Gordon & Breach Publishers, London.
2. Lang, S. B. 1966. Pyroelectric effect in bone and tendon. *Nature Lond.* **212**:704.
3. Athenstaedt, H. 1974. Pyroelectric and piezoelectric properties of vertebrates. *Ann. N.Y. Acad. Sci.* **238**:69.
4. Athenstaedt, H. 1976. Pyroelectric sensors of organisms. *Ferroelectrics.* **11**:365.
5. Athenstaedt, H. 1976. Pyroelectric properties of wheat. *Ferroelectrics.* **14**:753.
6. Athenstaedt, H. 1972. Pyroelectric behaviour of integument structures and of thermal-, photo-, and mechanoreceptors. *Z. Anat. Entwicklungsgesch.* **136**:249.

7. ATHENSTAEDT, H. 1972. Pyroelectric polarization in cells, tissues and organs of plants. *Z. Pflanzenphysiol.* **68**:82–91.
8. LIBOFF, A. R., and M. FURST. 1974. Pyroelectric effect in collagenous structures. *Ann. N.Y. Acad. Sci.* **238**:26.
9. FURST, M., and A. R. LIBOFF. 1977. Pyroelectric effect in collagenous materials, observed with pulsed 10.6 μm radiation. *Biophys. J.* **17**:300*a* (Abstr.).
10. LANG, S. B., and H. ATHENSTAEDT. 1978. Anomalous pyroelectric behavior in the leaves of the palm-like plant *Encephelartos villosus. Ferroelectrics.* **17**:511.
11. SIMHONY, M., and A. SHAULOV. 1971. Pyroelectric voltage response to step signals of infrared radiation in triglycine sulphate and strontium-barium niobate. *J. Appls. Phys.* **42**:3741.
12. SHAULOV, A., and M. SIMHONY. 1972. Pyroelectric voltage response to rectangular infrared signals in triglycine sulphate and strontium-barium niobate. *J. Appl. Phys.* **43**:1440.
13. SIMHONY, M., and A. SHAULOV. 1972. Measurement of the pyroelectric coefficient and permittivity from the pyroelectric response to step radiation signals in ferroelectrics. *Appl. Phys. Lett.* **21**:375.
14. LINES, M. E., and A. M. GLASS. 1977. Principles and Applications of Ferroelectrics and Related Materials. Oxford University Press, Oxford. 569.