POLYMERIC SPACE-CHARGE ELECTRETS

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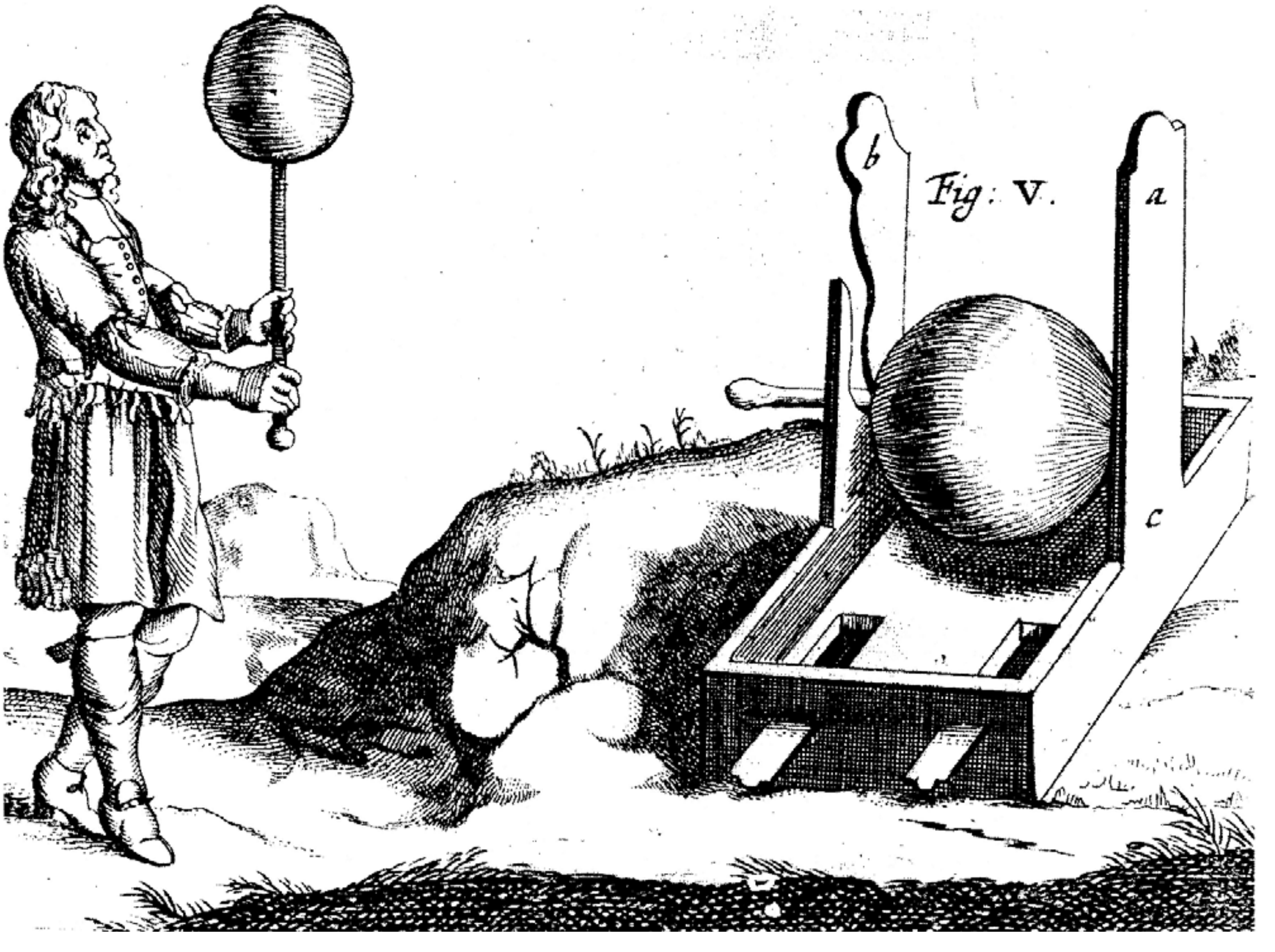
1. A little bit of history and some introduction:
   Electrophore or space-charge electret?
2. Charge-storing polymers for electret applications:
   Typical materials and essential requirements
3. Acoustical and thermal charge-probing techniques:
   Looking inside without cutting the polymer open
4. The mystery of charge-trapping mechanisms:
   Chemical trapping, physical trapping or both?
5. Conclusions and outlook


b
Fig: V.
a
c

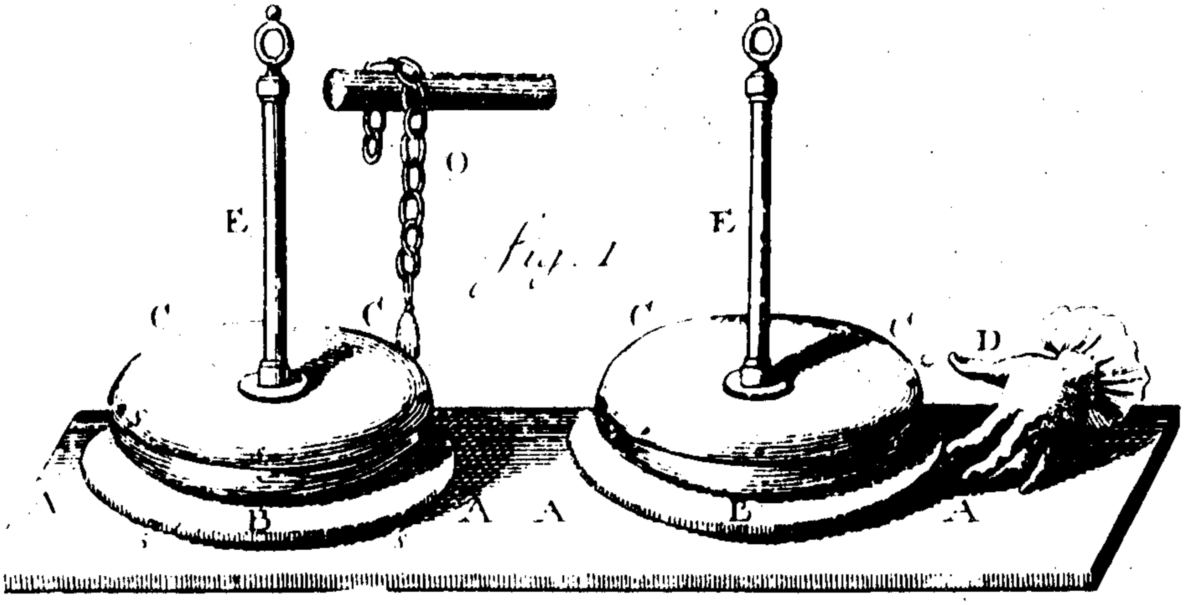


fig. 1

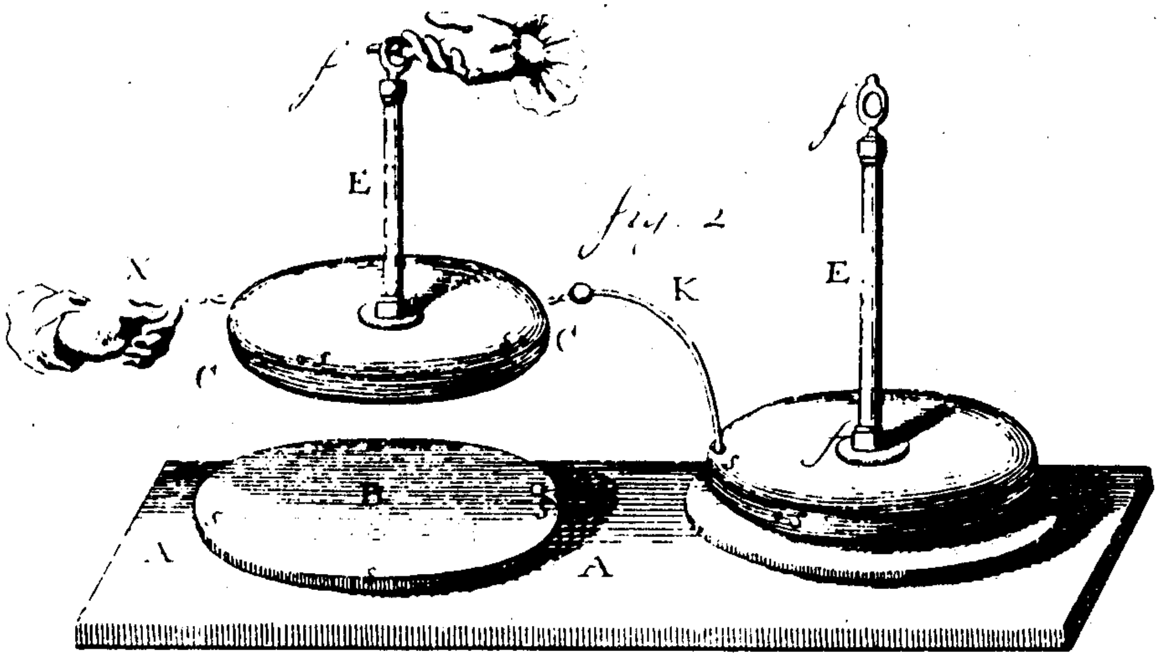


fig. 2

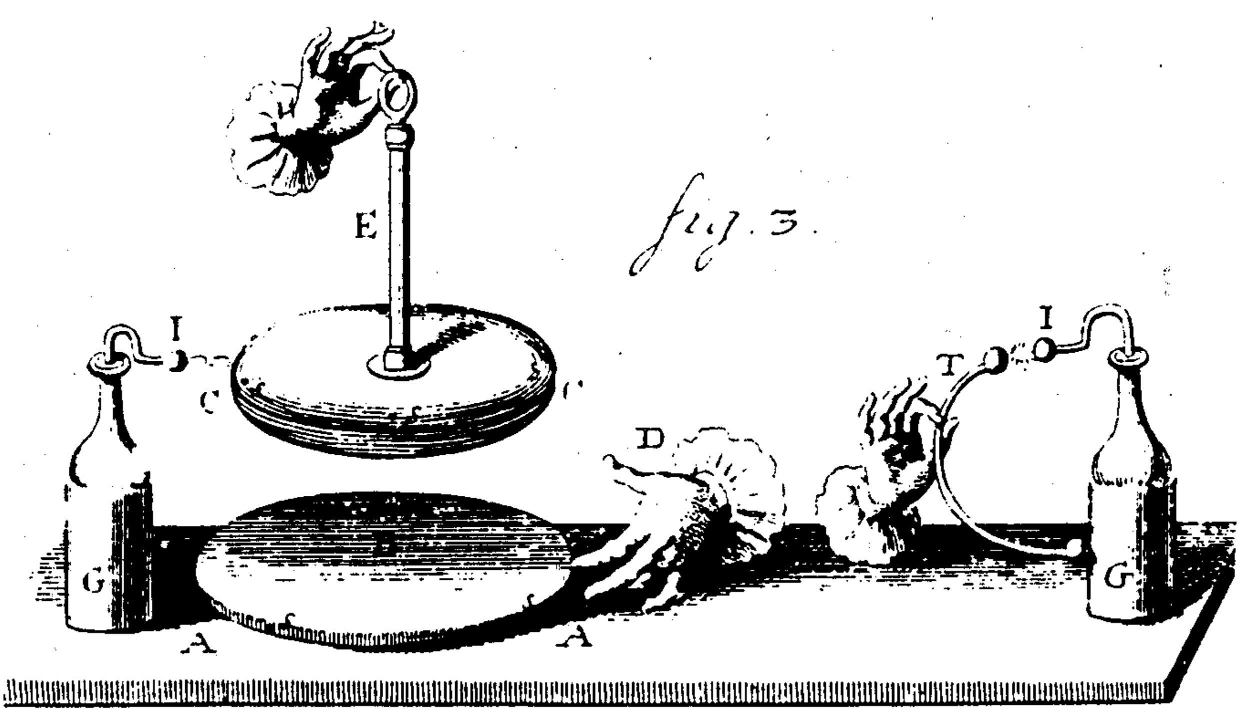
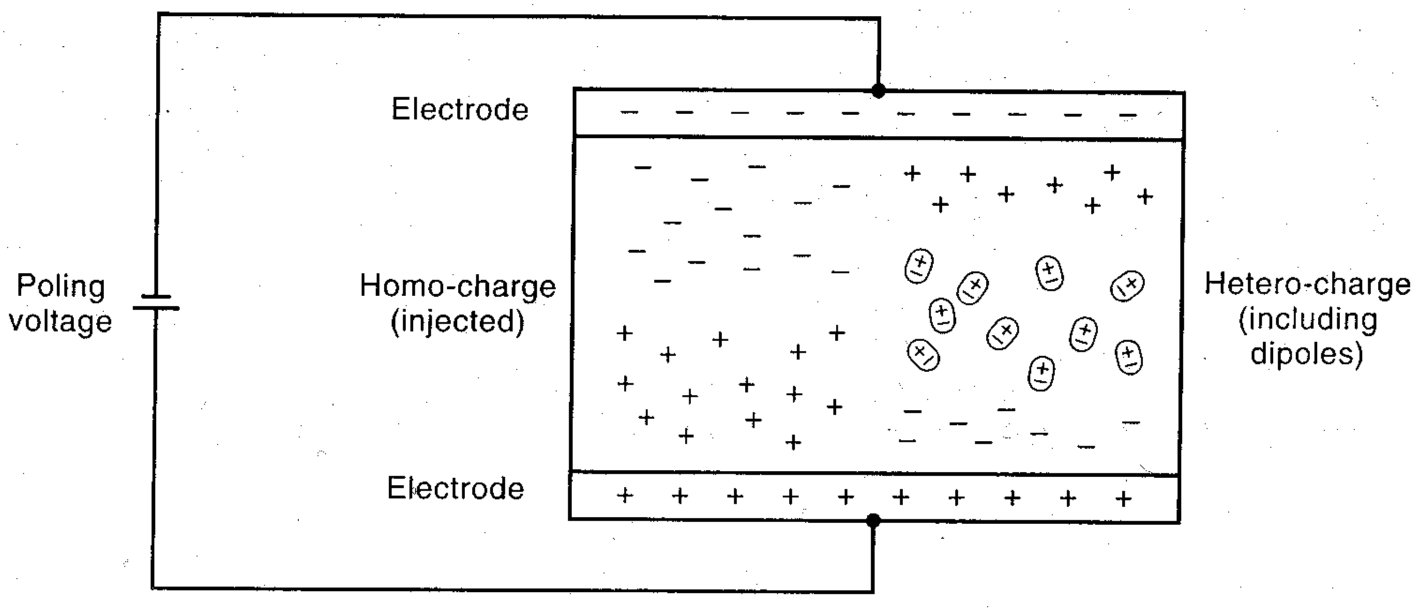


fig. 3

- Thales of Miletus (~640–546 B.C.): Knowledge about the attractive force of magnets and amber
- William Gilbert (1544–1603): Clear distinction between "magnets" and (di-)"electrics"
- Otto von Guericke (1602–1686): Electrification of sulfur and demonstration of electric forces
- Stephen Gray (1666–1736): Discovery of several electret materials and of electrical conduction
- Petrus van Musschenbroek (1692–1761) and Ewald Georg von Kleist (~1700–1748): Discovery of the "Leiden" or "Kleist" jar (glass capacitor)
- Benjamin Franklin (1706–1790): Theory of the "electrical fire" (including charge conservation)
- Franz Ulrich Theodosius Aepinus (1724–1802): Qualitative theory of electro- and magnetostatics
- Johan Carl Wilcke (1732–1796) and Alessandro Volta (1745–1827): Invention of "electrophorus"
- Charles-Augustin Coulomb (1736–1806): Law of the forces between electric charges on bodies
- Oliver Heaviside (1850–1925): First proposal of the name "electret" (in analogy to "magnet")

(Injected) homocharge (left) and (separated) **heterocharge** (right) in a polymer electret

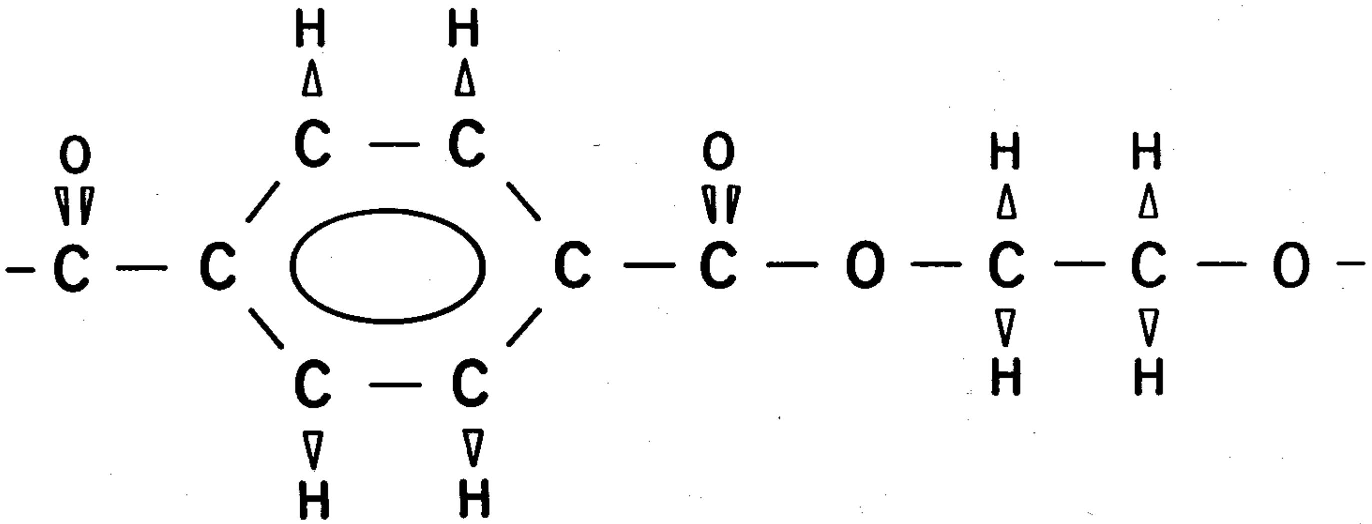


Typical space-charge electret polymers (fluoroethylenepropylene copolymer, polyimide, polyethyleneterephthalate, cycloolefin copolymer, polypropylene) and some properties

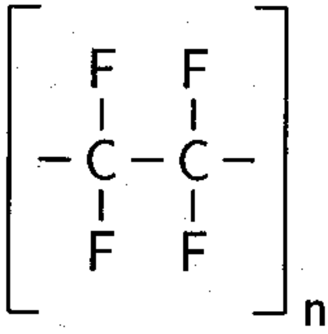
Name	Acronym	Density	Melting/Degrad.	Permittivity	Resistivity	Elasticity
Teflon	FEP	2.15 Mg/m^3	$\approx$ 270 °C	2.0 ε_0	$> 10^{16}\,\Omega$m	480 MPa
Kapton	PI	1.42 Mg/m^3	(>400 °C)	>3.0 ε_0	$> 10^{15}\,\Omega$m	2.5 GPa
Mylar	PETP	1.39 Mg/m^3	$\approx$ 255 °C	3.3 ε_0	$\approx 10^{16}\,\Omega$m	4.8 GPa
Topas	COC	1.02 Mg/m^3	$\approx$ 260 °C	2.35 ε_0	$> 10^{14}\,\Omega$m	3.2 GPa
many	PP	0.91 Mg/m^3	$\approx$ 165 °C	2.2 ε_0	$> 10^{16}\,\Omega$m	1.8 GPa

Note: Names are usually protected as registered trademarks and vary from manufacturer to manufacturer. Only one typical trade name for each electret polymer is given here.

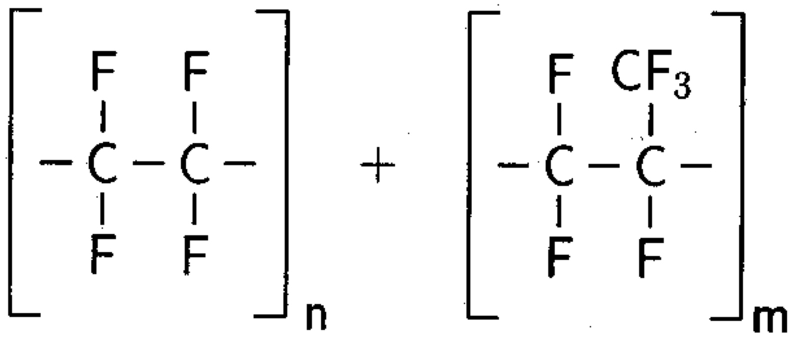
Chemical formula of polyethylene terephthalate (PETP or PET)
Trade names: Mylar, Hostaphan, Terphane, Melinex, etc.



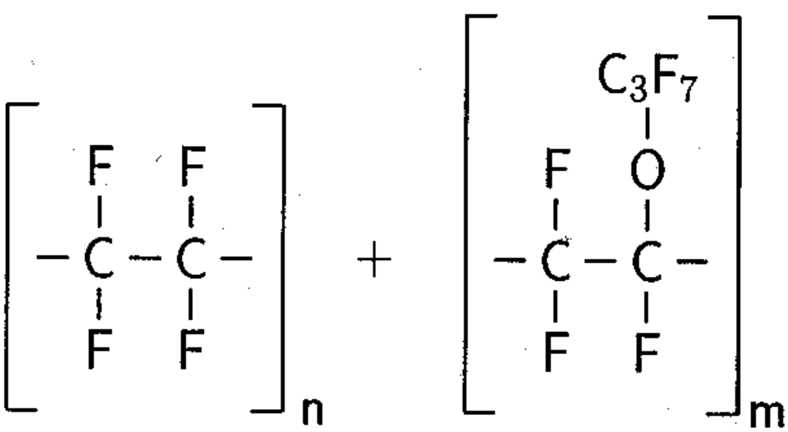
Teflon Fluoropolymers for Space-Charge Electrets



Polytetrafluoroethylene (Teflon PTFE)

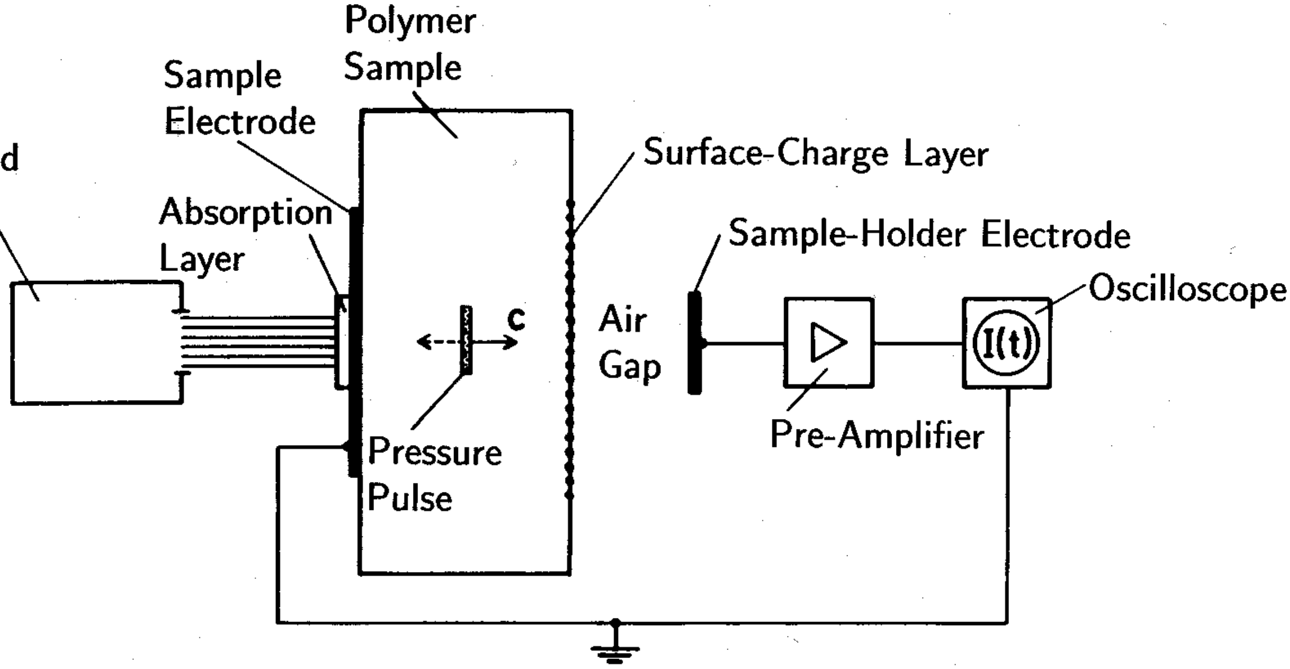


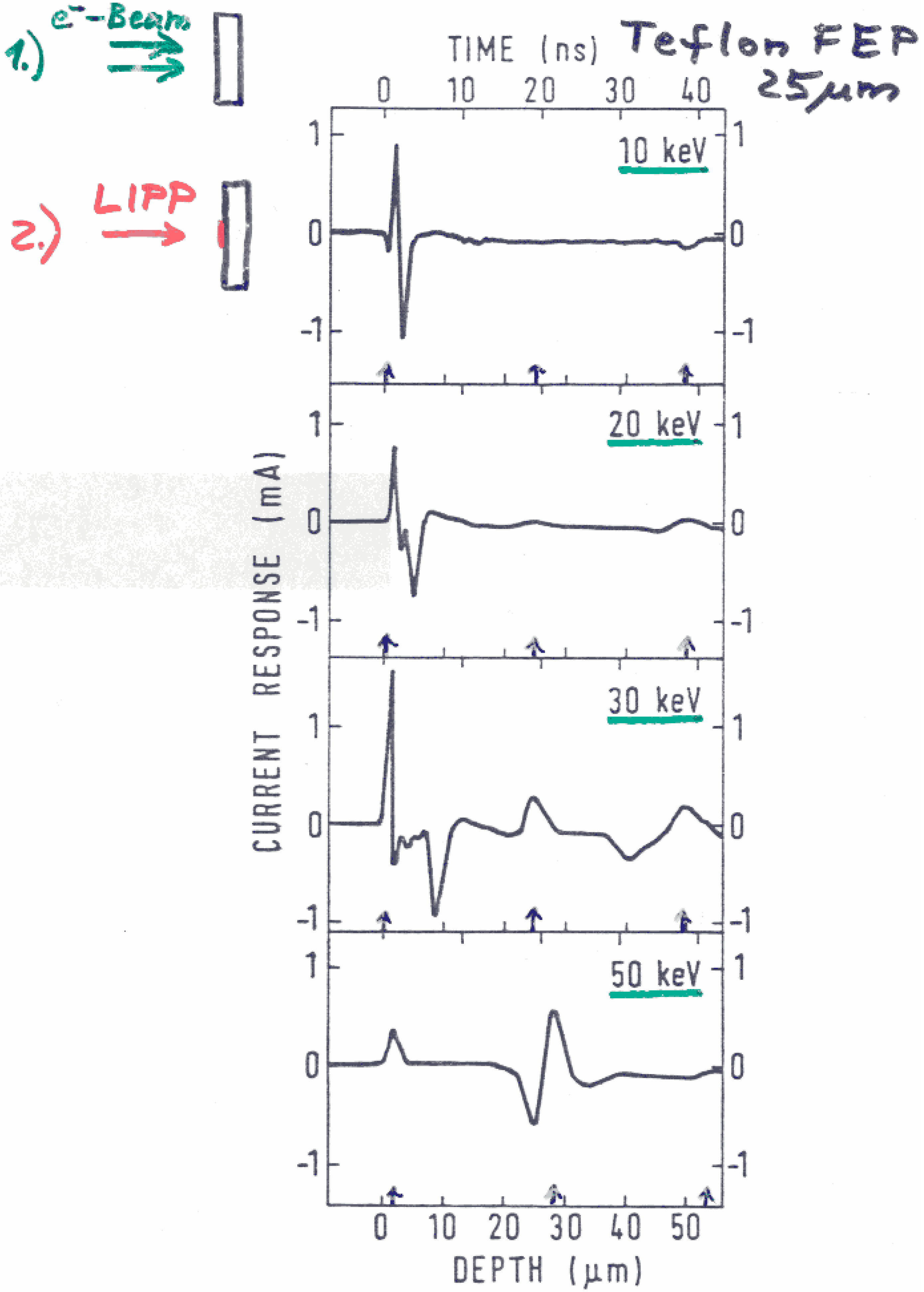
Tetrafluoroethylene-co-hexafluoropropylene
(Fluoroethylenepropylene, Teflon FEP)



Tetrafluoroethylene-co-perfluoropropoxyethylene
(Perfluoroalkoxy, Teflon PFA)

Laser-Induced Pressure Pulses (LIPPs) for probing charge and polarization-gradient profiles in polymer films

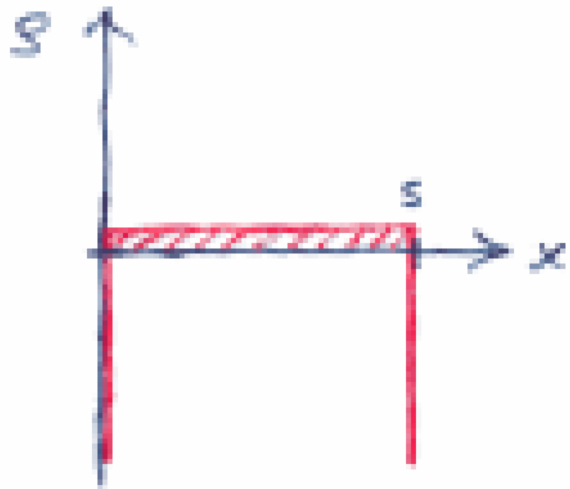




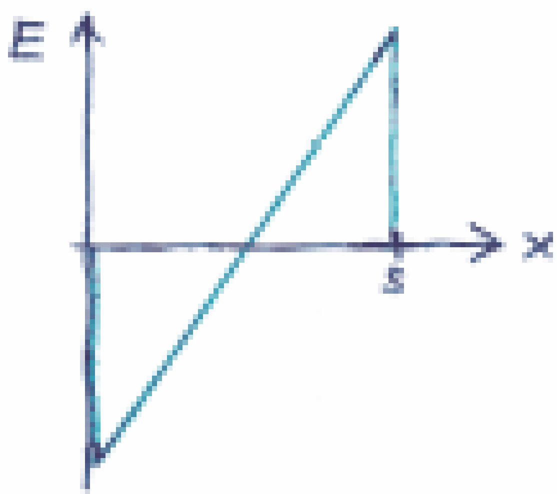
1.) e⁻-Beam
2.) LIPP
TIME (ns)
Teflon FEP 25µm
10 keV
20 keV
30 keV
50 keV
CURRENT RESPONSE (mA)
DEPTH (µm)

Uniform Volume-Charge Distributions

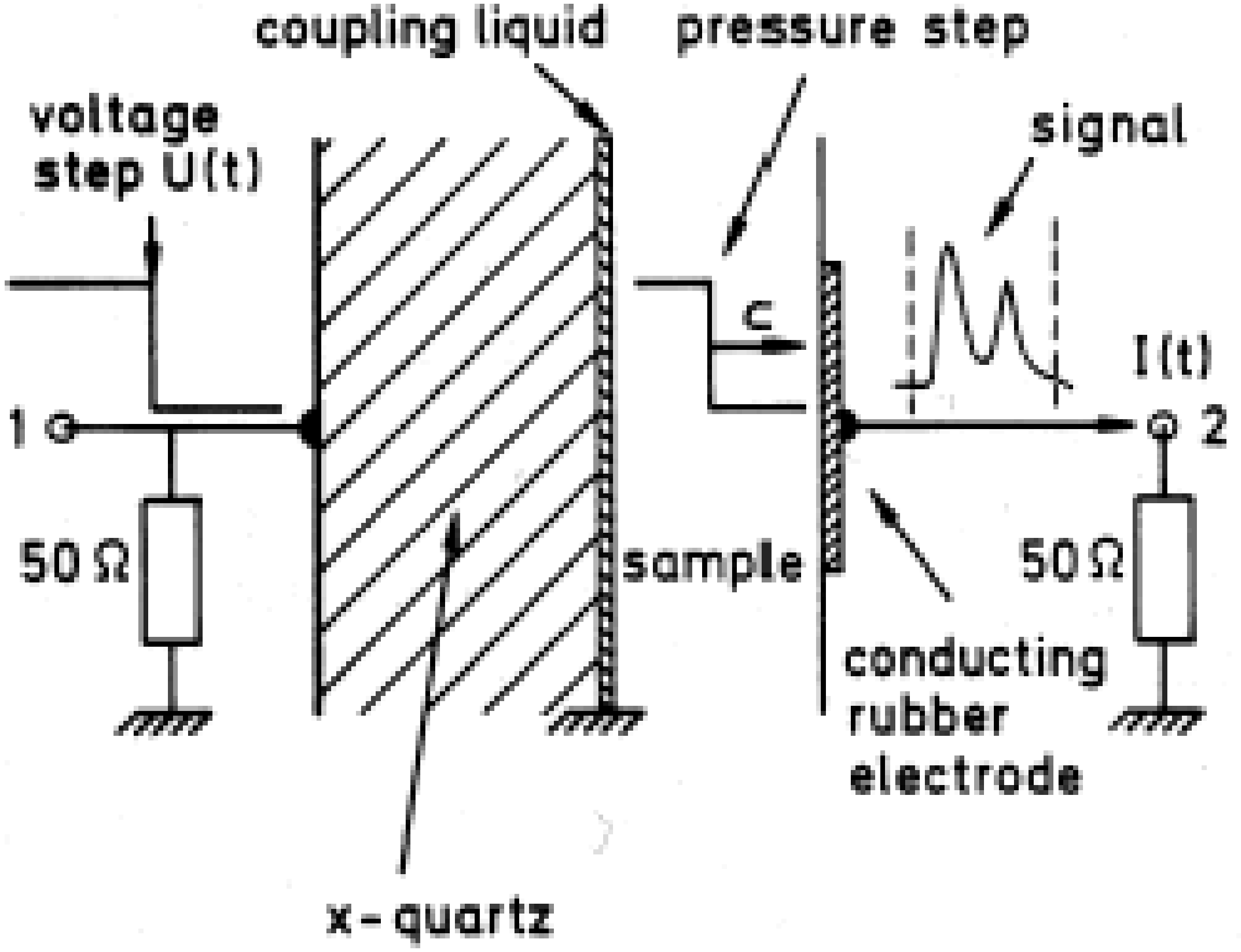
- in Pressure-Pulse Experiments
  ($I(t) \sim \rho(x)$; e.g. LIPP)



- in Pressure-Step Experiments
  ($I(t) \sim E(x) \sim \int \rho(x)\,dx$; e.g. PPS)

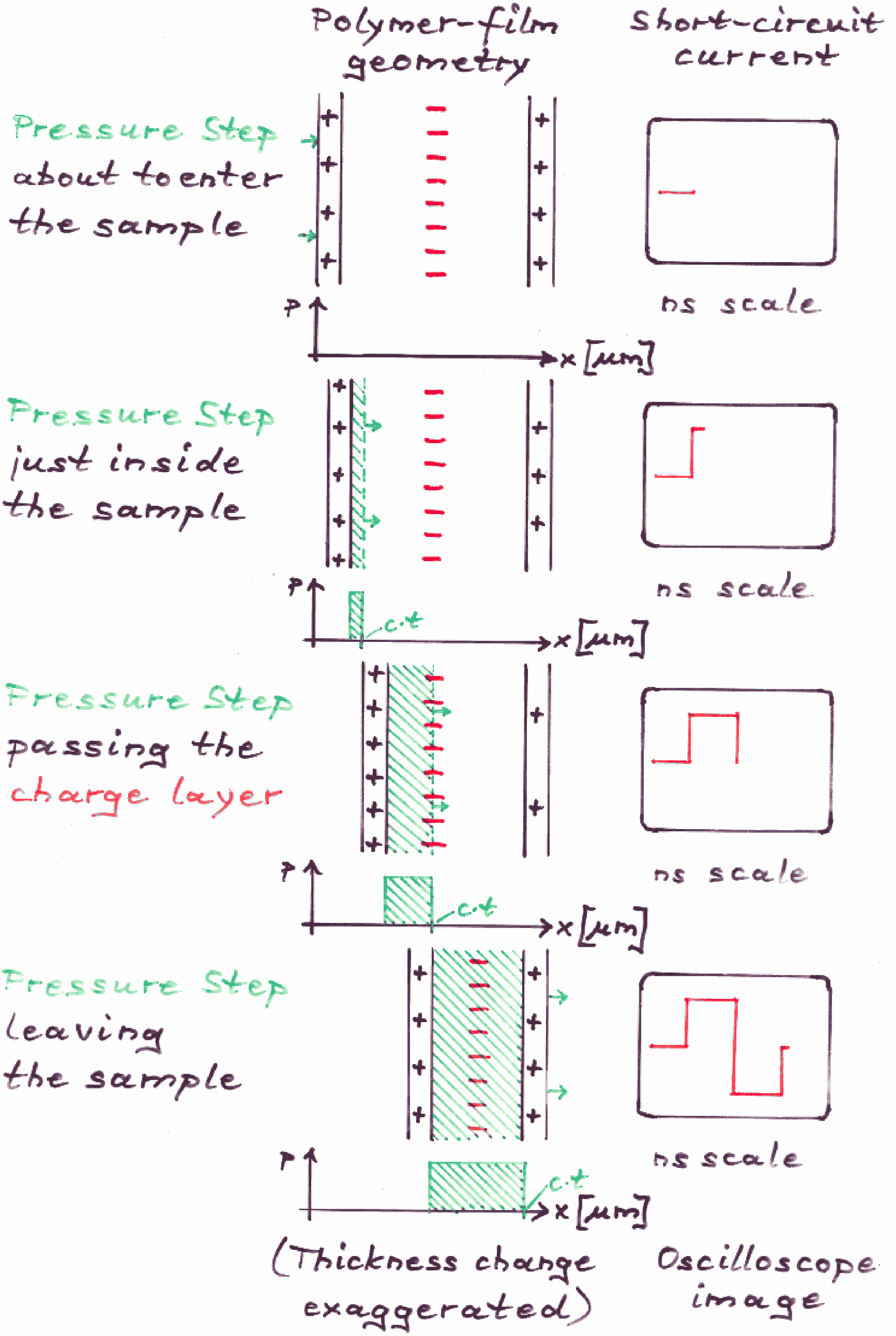


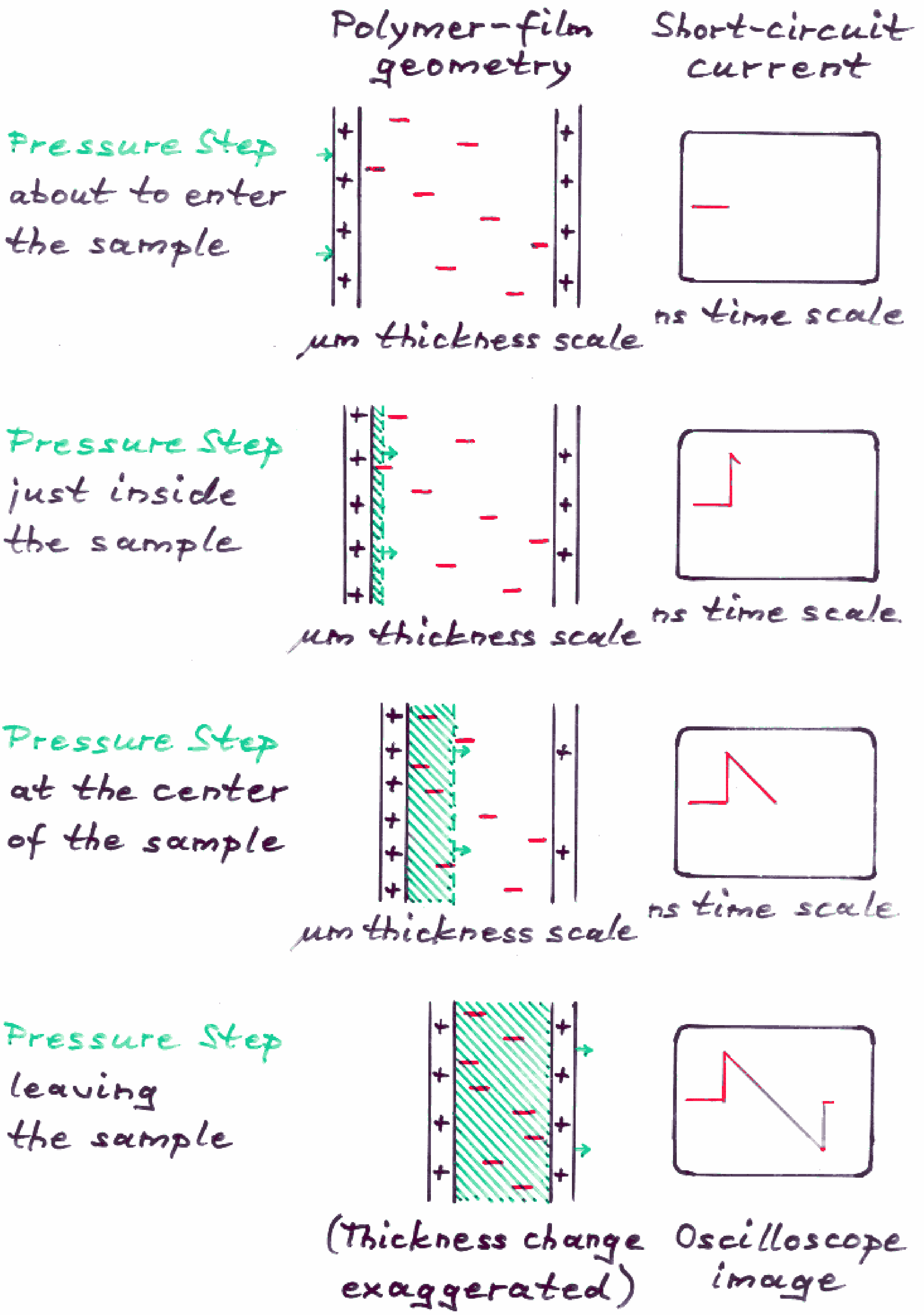
Piezoelectrically Generated Pressure Steps for the Detection of Electric-Field Profiles



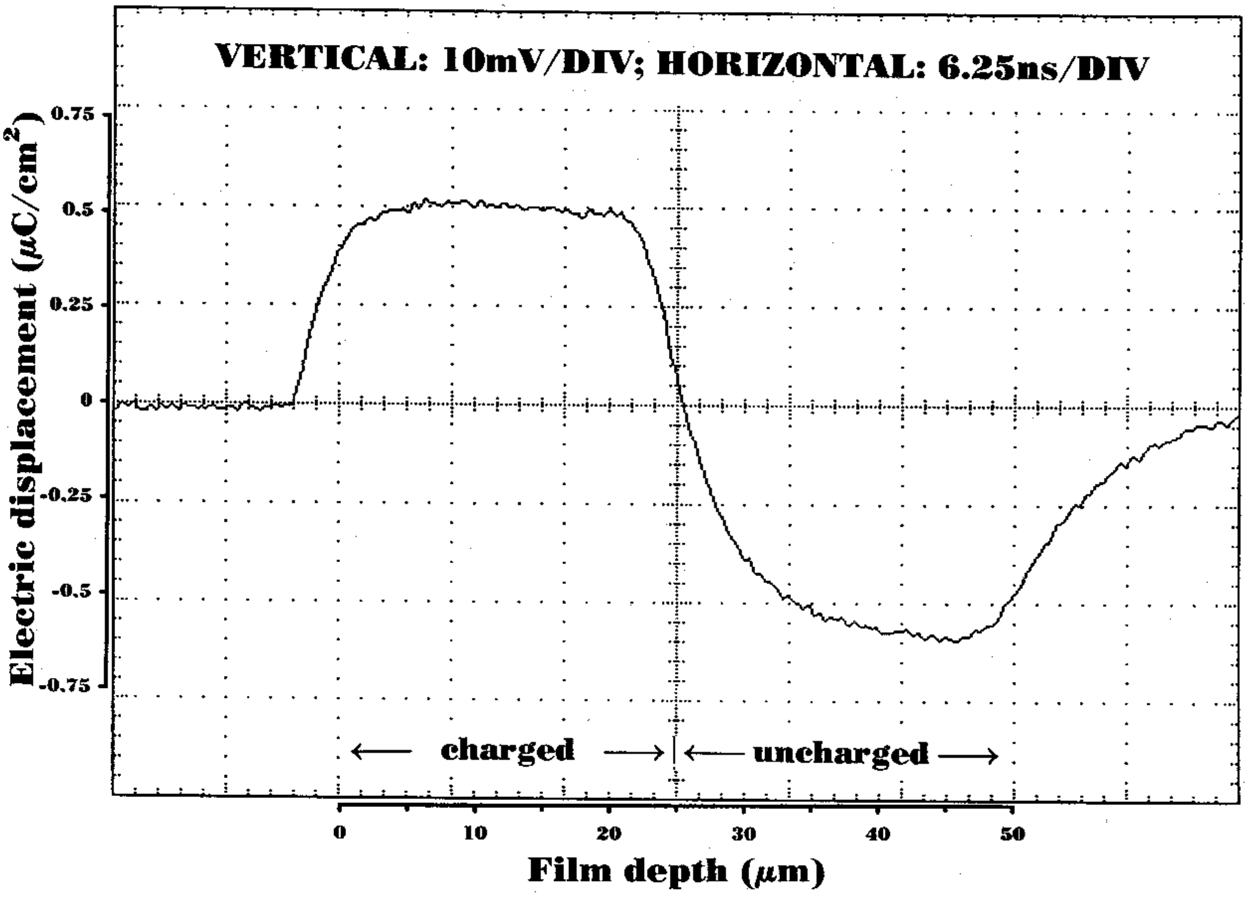
$$I(t) = \frac{Ap}{\rho_0 cs}(\gamma+1)\int_0^{x=ct}\rho(\xi)d\xi$$

$$= \frac{Ap}{\rho_0 cs}(\gamma+1)\varepsilon_0\varepsilon\, E(x=ct)$$

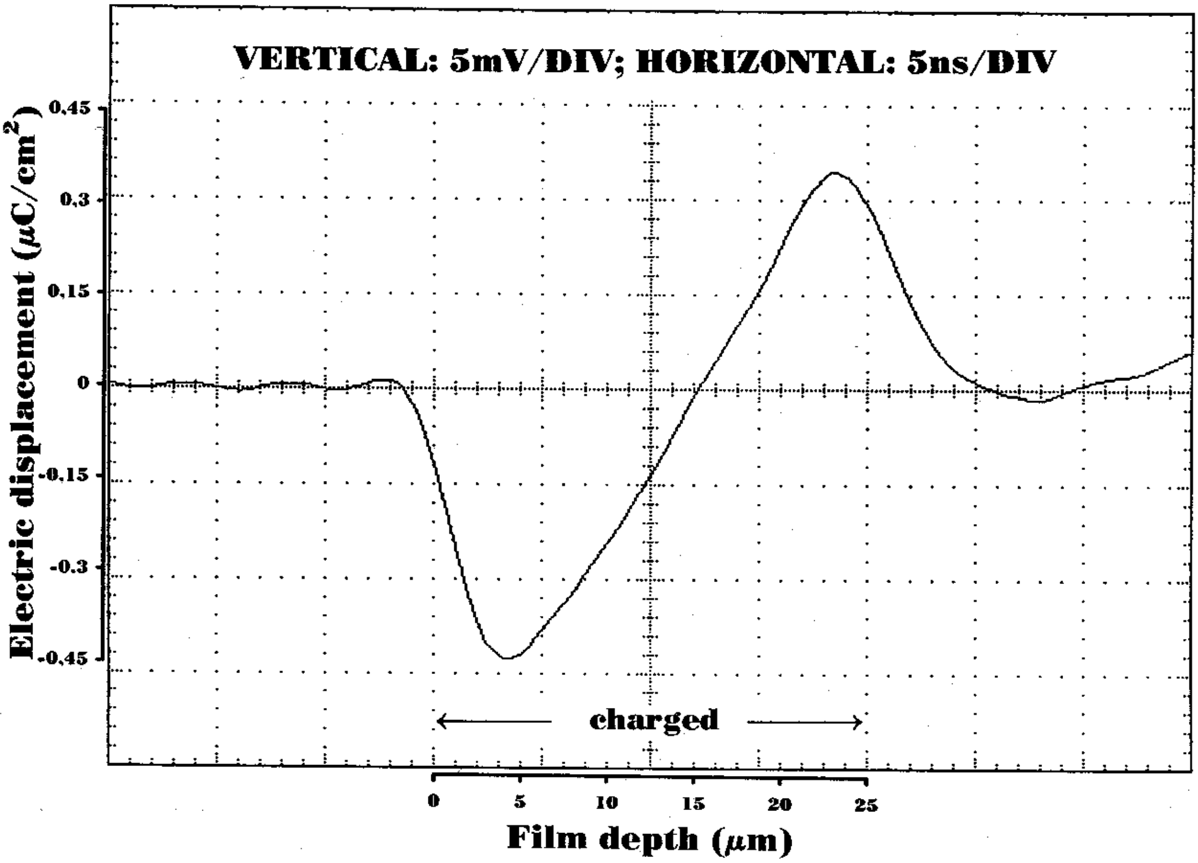

Polymer-film geometry
Short-circuit current
Pressure Step about to enter the sample
ns scale
p
x [µm]
Pressure Step just inside the sample
c·t
ns scale
p
x [µm]
Pressure Step passing the charge layer
c·t
ns scale
p
x [µm]
Pressure Step leaving the sample
c·t
ns scale
p
x [µm]
(Thickness change exaggerated)
Oscilloscope image



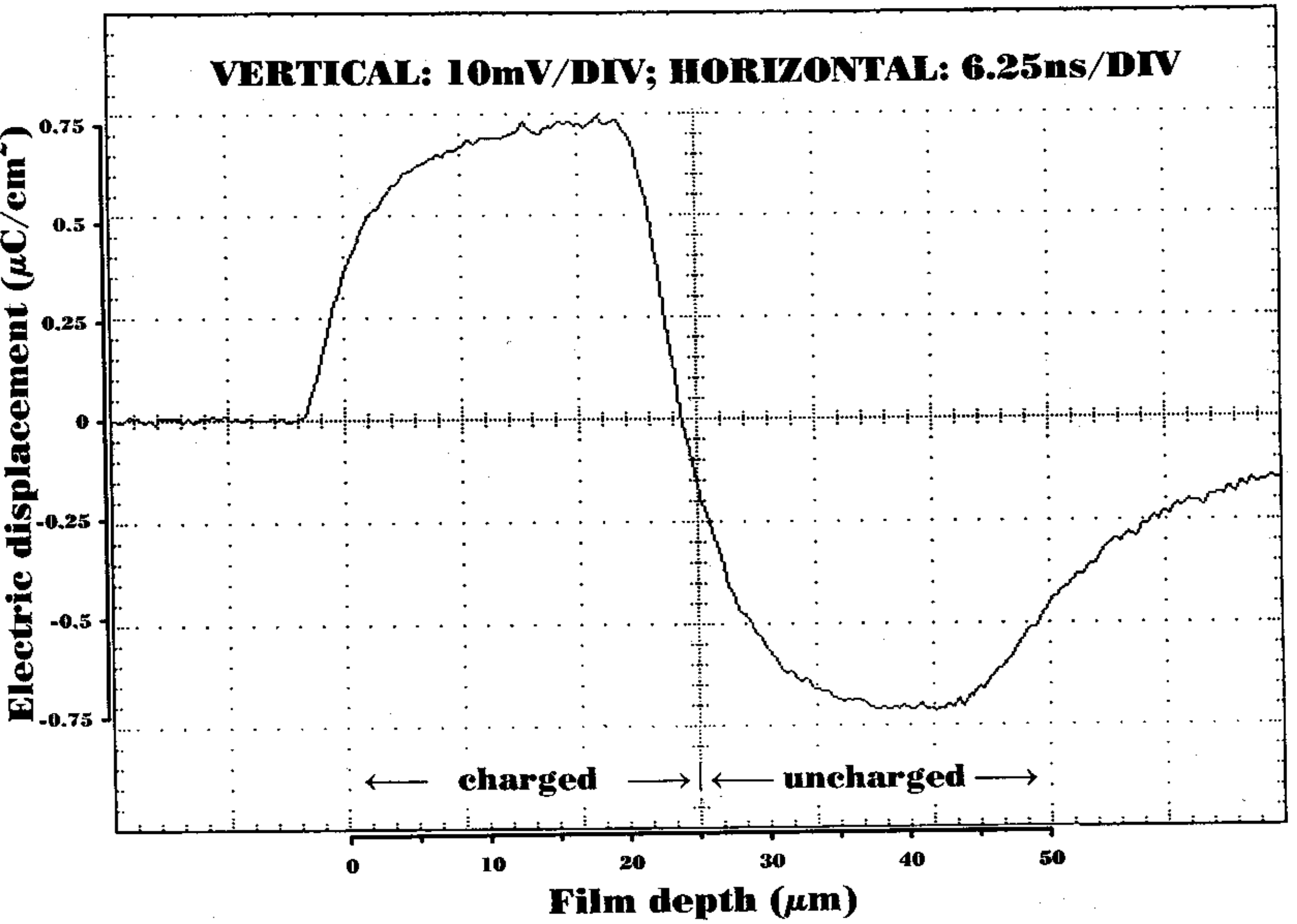
Polymer-film geometry
Short-circuit current
Pressure Step about to enter the sample
µm thickness scale
ns time scale
Pressure Step just inside the sample
µm thickness scale
ns time scale
Pressure Step at the center of the sample
µm thickness scale
ns time scale
Pressure Step leaving the sample
(Thickness change exaggerated)
Oscilloscope image



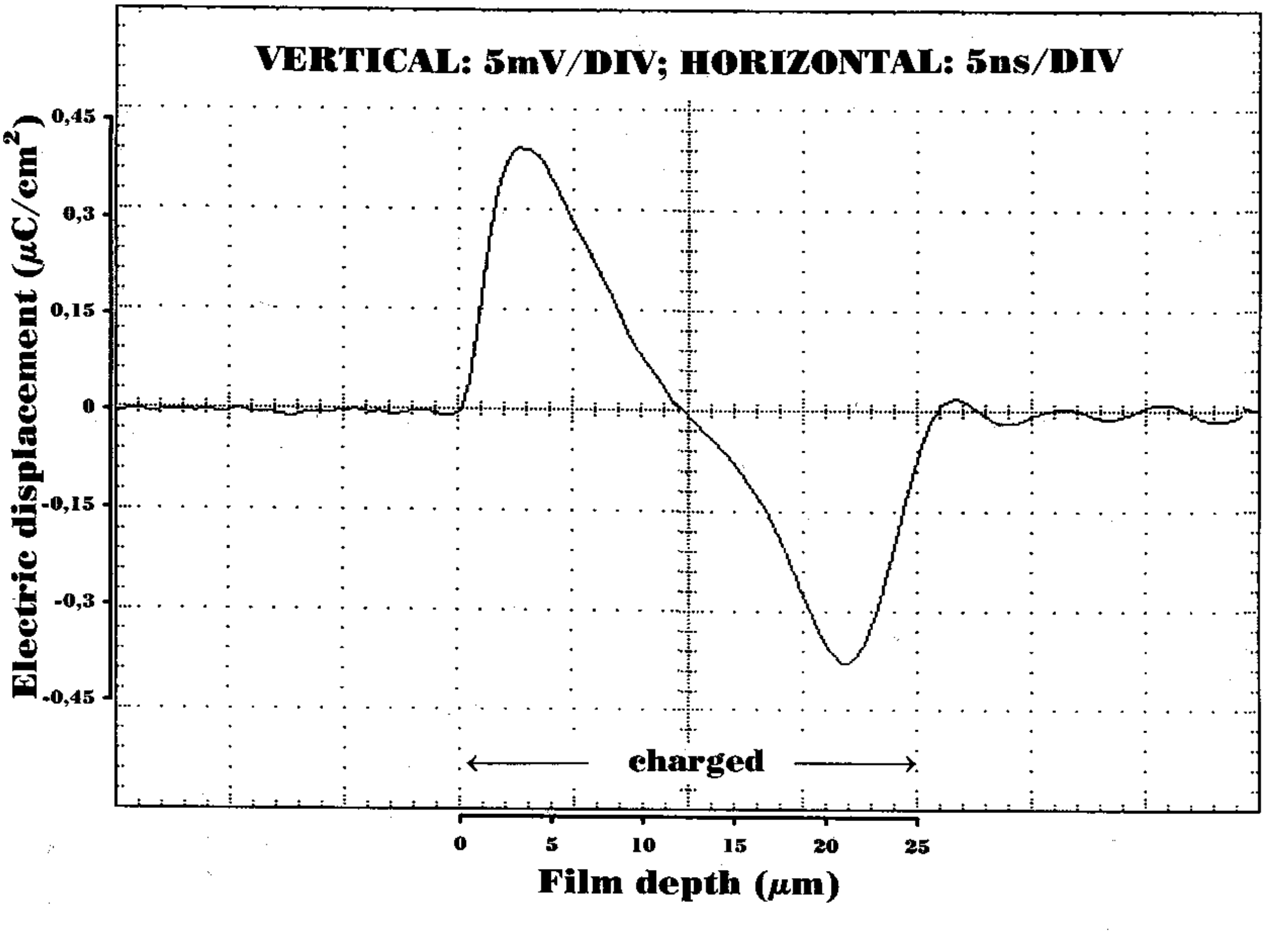
Electric-displacement profile in negatively corona-charged Teflon FEP (V_C=-15 kV, T_C=20°C, t_C=60 min, maximum charge density ≈ 1600 C/m³)



Electric-displacement profile in positively corona-charged Teflon PFA (V_C=-15 kV, T_C=200°C, t_C=60 min, maximum charge density ≈ 500 C/m^3)



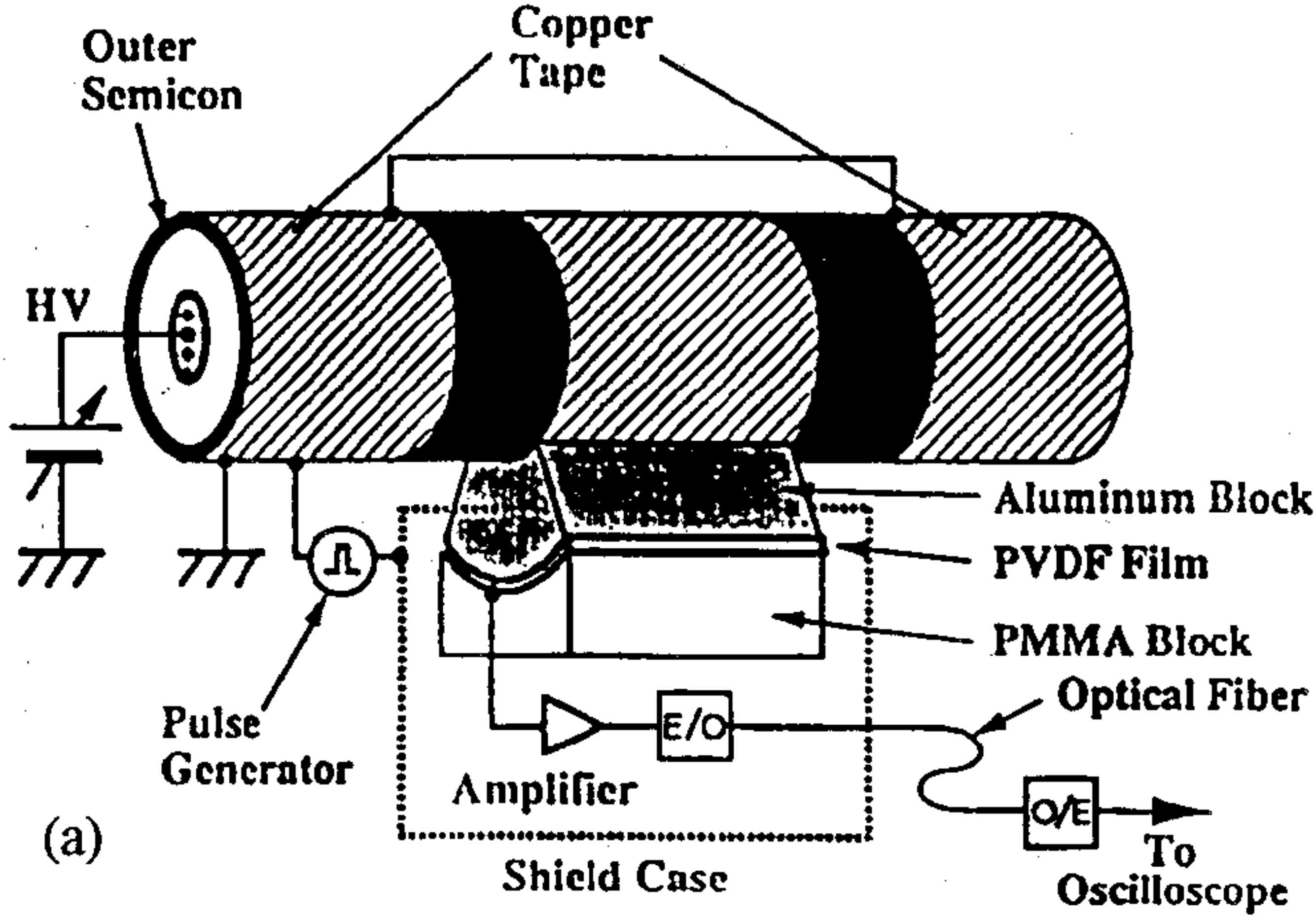
Electric-displacement profile in negatively corona-charged Teflon PFA (V_C=-15 kV, T_C=20°C, t_C=60 min, maximum charge density ≈ 2000 C/m³)



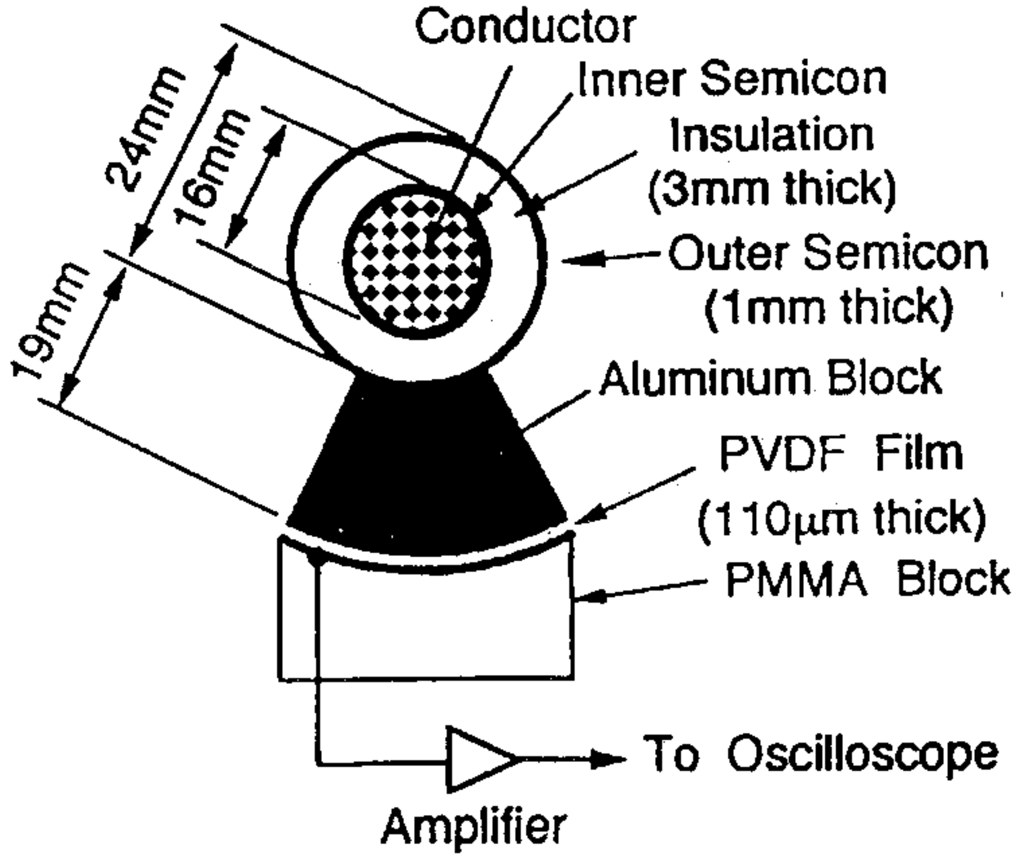
Electric-displacement profile in negatively corona-charged Teflon FEP (V_C=-15 kV, T_C=200°C, t_C=60 min, maximum charge density ≈ 500 C/m^3)

Side view (a) and cross section (b) of special PEA equipment for *in-situ* measurements on HV cables

(after Hozumi *et alii* 1994)



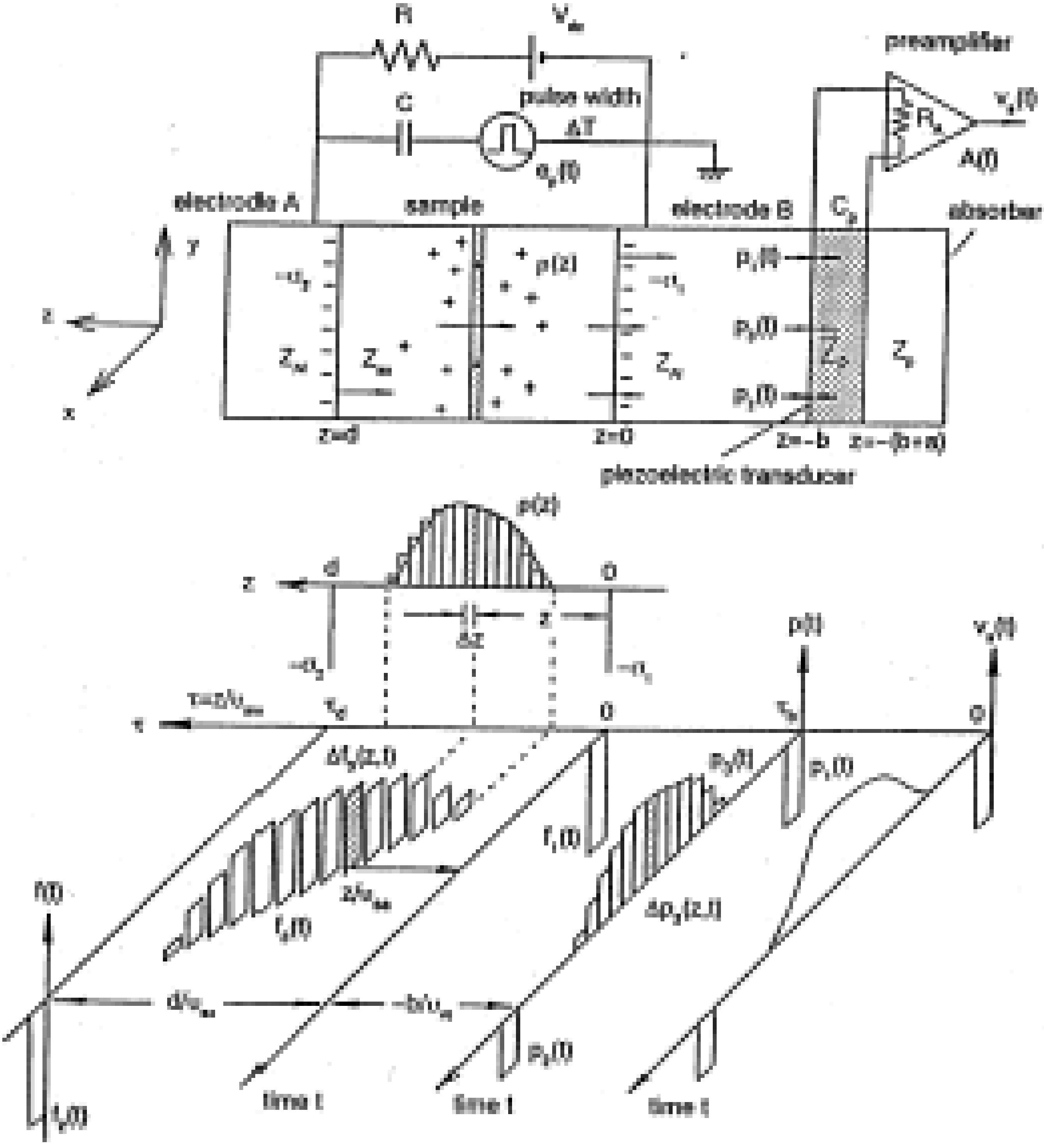
(a)



(b)

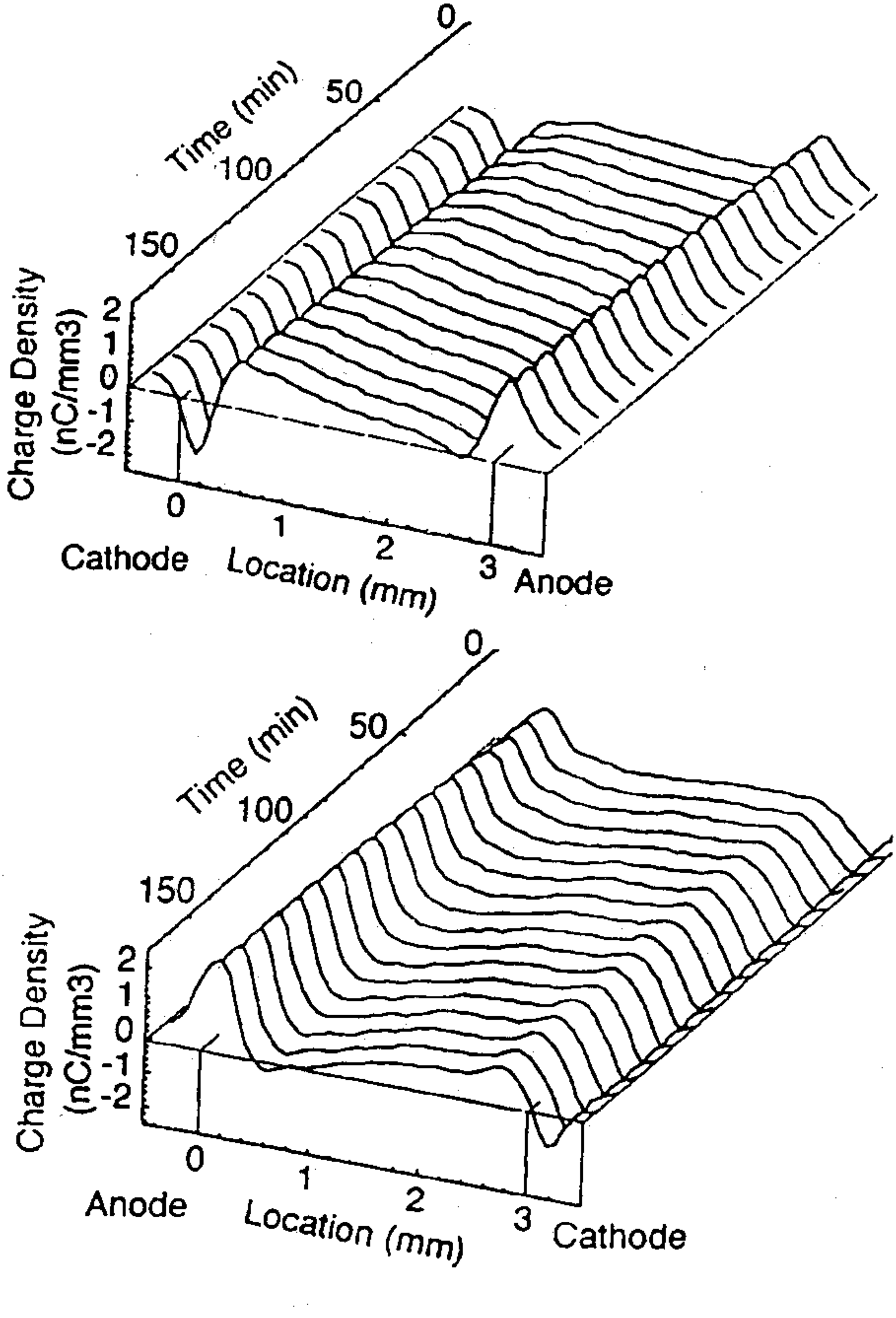
"Pulsed electroacoustic" (PEA) method for the measurement of charge distributions in dielectrics

(after Takada and Li 1992)



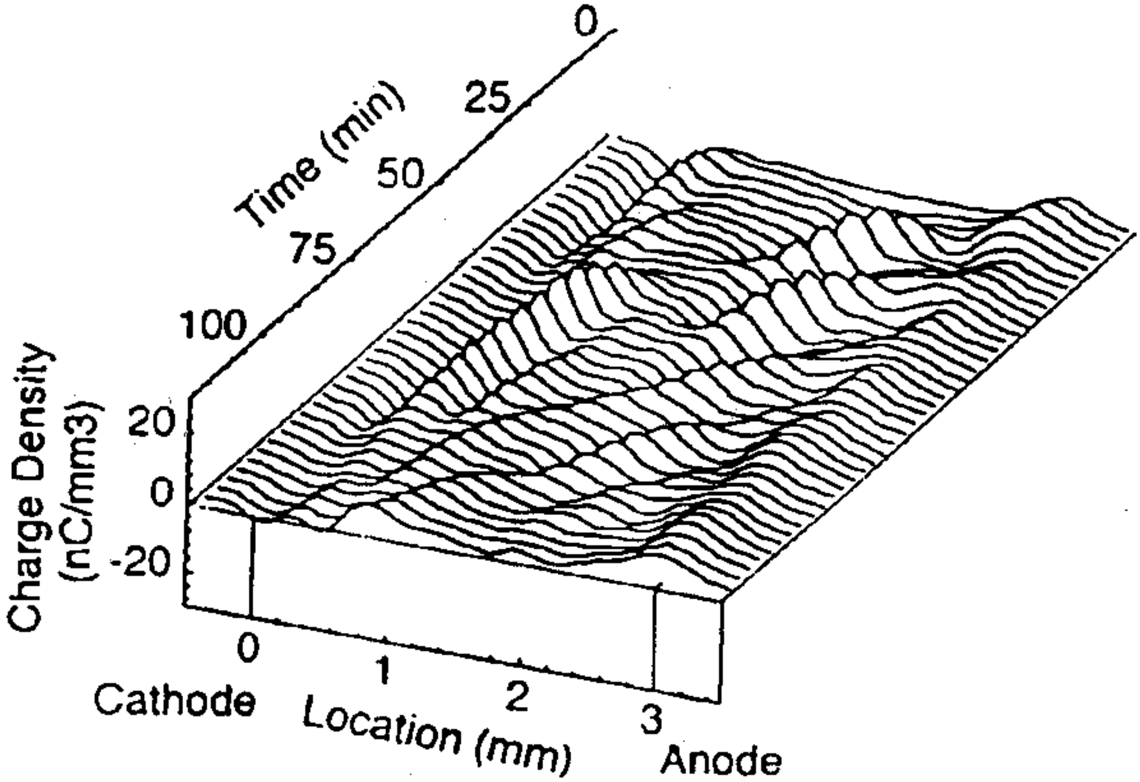
Development of the space-charge distributions in a high-voltage cable during application of ±70kV

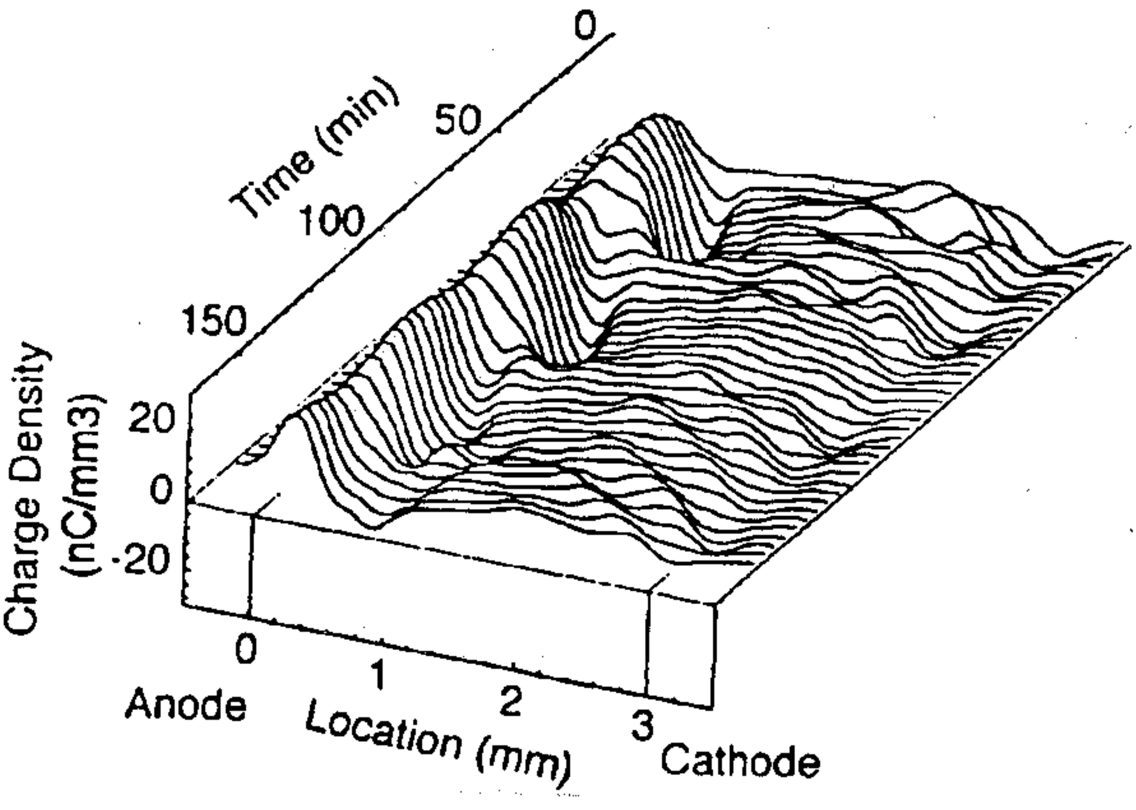
(after Hozumi *et alii* 1994)



Development of the space-charge distributions in a high-voltage cable during application of ±350kV

(after Hozumi *et alii* 1994)





The 6 most relevant schemes for non-destructive probing of space charge in electret films

Probing technique	Mechanism of operation	Excitation mechanisms	Detection of signal	Spatial resolution
Thermal pulse	Time-dependent non-uniform expansion	Light flash or pulsed laser	Voltage with oscilloscope	depends on depth
Thermal step	Time-dependent non-uniform expansion	Heat reservoirs or continous laser	Voltage with oscilloscope	depends on depth
Thermal wave	Frequency-dependent periodic expansion	Chopped light or modulated laser	Current/voltage with lock-in amp.	depends on freq.
Acoustic pulse	Propagation of thin compression zone	Piezo-transducer or pulsed laser	Current via 50Ω with oscilloscope	$\geq 1\mu$m
Acoustic step	Propagation of steep compression front	Cable discharge + piezo-transducer	Current via 50Ω with oscilloscope	$\geq 1\mu$m
Acoustic wave	Propagation of field-induced acoustic wave	Electric pulse on electrodes	Piezo-transducer + oscilloscope	depends on pulse

Response Equations for the Probing of Charge and Polarization Profiles in Thin Dielectrics by Non-Uniform Temperature or Stress Excitation

Total current density inside the sample:

$$i(t) = \rho_r(x,t)v(x,t) + \frac{\partial D(x,t)}{\partial t} \quad \text{with} \quad D(x,t) = \varepsilon_0 \varepsilon(x,t)E(x,t) + P(x,t)$$

General response equation (T ≡ temperature or stress):

$$I(t) = \frac{A}{s}\int_0^s \left[\left(\frac{1}{\varepsilon}\frac{d\varepsilon}{dT} - \frac{1}{x}\frac{dx}{dT}\right)\varepsilon_0\varepsilon E(x) + \frac{dP(x)}{dT}\right]\frac{\partial T(x,t)}{\partial t}dx$$

Response equation for thermal excitation (e.g. heat pulse):

$$I(t) = \frac{A}{s}\int_0^s [(\alpha_\varepsilon - \alpha_x)\varepsilon_0\varepsilon E(x) + \lambda(x)]\frac{\partial T(x,t)}{\partial t}dx$$

Response equation for mechanical excitation (e.g. pressure pulse):

$$I(t) = \frac{A}{s}\int_0^s [-(\gamma+1)\varepsilon_0\varepsilon E(x) + e_{33}(x)]\chi\frac{\partial p(x,t)}{\partial t}dx$$

Response Equations for Specific Experimental Probing Techniques

Laser-Intensity Modulation Method (LIMM) (Lang and Das-Gupta 1981)

$$I(t) = \frac{A}{s} \int_0^s \left[(\alpha_\varepsilon - \alpha_x) \int_0^x \rho_r(\xi) d\xi + (\alpha_P + \alpha_x - \alpha_\varepsilon) P(x) \right] \frac{\partial T(x,t)}{\partial t} dx$$

Thermal Pulsing Technique (Collins 1975)

$$\Delta V(t) = \int_0^s \left[\frac{\alpha_\varepsilon - \alpha_x}{\varepsilon_0 \varepsilon} \int_0^x \rho_r(\xi) d\xi + \frac{\alpha_P + \alpha_x - \alpha_\varepsilon}{\varepsilon_0 \varepsilon} P(x) \right] \Delta T(x,t) dx$$

Laser-Induced Pressure-Pulse (LIPP) Technique
(Sessler, West and Gerhard-Multhaupt 1981)

$$I(t) = \frac{Ap\tau}{\rho_0 s} \left[(\gamma + 1) \varepsilon_0 \varepsilon \rho(x) - \frac{de_{33}(x)}{dx} \right]_{x=ct}$$

Piezoelectrically Generated Pressure-Step (PPS) Method
(Eisenmenger and Haardt 1982)

$$I(t) = \frac{Ap}{\rho_0 c s} \left[(\gamma + 1) \int_0^x \rho(\xi) d\xi - e_{33}(x) \right]_{x=ct}$$

- Space charge (i.e. *extra* charge) provides external electric field for applications
- Good charge retention related to excellent insulation properties and high thermal stability
- A few special polymers (plus some inorganic materials) highly suitable for electrets

- Non-destructive space-charge probing with acoustic-wave (pressure) propagation or thermal-wave (heat) diffusion possible
- Space-charge stability possibly a combination of deep (chemical) trapping on macromolecule and restricted (physical) mobility of macromolecules