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Modelling charge generation and transport in solid organic dielectrics under DC stress

S. LE ROY, T. T. Nga VU, C. LAURENT, G. TEYSSEDRE

Overview

- Context
- Existing types of models in solid dielectrics
- Intermediate models:
  - Identification of physical hypotheses
  - Space charge in a cable geometry: simulation results

Polymeric HVDC technology

- XLPE as electrical insulation for HVDC transmissions
  - HVDC LCC
  - HVDC VSC

- ADVANTAGES OF HVDC:
  - Transport of electricity over long distances
  - Link with networks of different frequencies

- HVDC APPLICATIONS

→ RELIABILITY

Industrial Background

RELIABILITY ?

Problem: presence of electrical charges

Electric field enhancement

Aging

dielectric breakdown

MODELS

To understand the causes and consequences of the presence of charges in solid dielectrics

Overview

- Context
- Existing types of models in solid dielectrics
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  - Space charge in a cable geometry: simulation results

Existing models in solid dielectrics

- **Microscopic models:**
  + energy levels linked to chemical or physical defects
  + Density Functional Theory (DFT) calculations – quantuum mechanical computations

☺ rigorous description

☹ high calculation time

☹ no prediction of the macroscopic characteristics

- **Macroscopic models:**
  + conductivity / permittivity as a function of field and temperature
  + based on experiments

- **Semi-microscopic – Intermediate - models:**
  + transport phenomena
  + microscopic phenomena with effective parameters

Examples of results from microscopic models

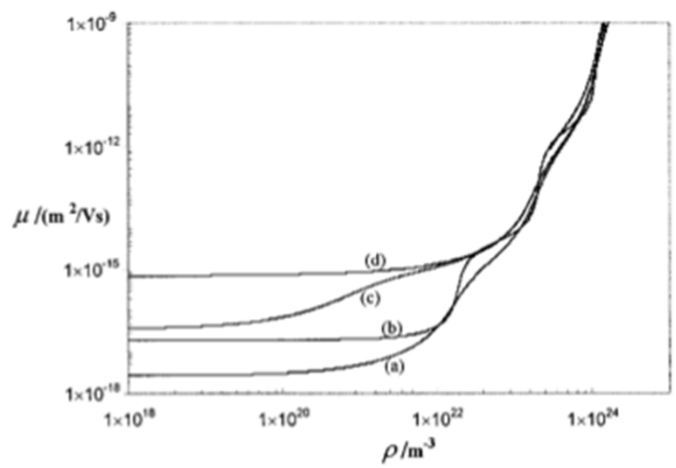
M. Meunier, N. Quirke, et al

Calculation of the trap depth as a function of the chemical impurities in XLPE [1]

Molecule	Electron affinity	Trap depth
(a) decane (n-$C_{10}H_{22}$)	−1.366[a]	0
(b) 5-decanone ($C_{10}H_{20}O$)	−0.913	0.453
(c) 5-decene ($C_{10}H_{20}$)	−1.244	0.122
(d) 4,6-decene ($C_{10}H_{18}$)	−0.923	0.443
(e) 5-decyne ($C_{10}H_{18}$)	−1.325	0.041
(f) 5-vinyl nonane ($C_{10}H_{20}$)	−1.209	0.157
(g) 5-decanol ($C_{10}H_{21}O$)	−1.180	0.186
(h) 5-decanal ($C_{10}H_{20}O$)	−0.921	0.445
(i) 4-propyl heptane ($C_{10}H_{22}$)	−1.245	0.121
(j) alpha-methylstyrene, C_9H_{10}	0.23	1.53
(k) Cumylalcohol, $C_9H_{12}O$	−1.02	0.28
(l) Acetophenone, C_8H_8O	−0.40	0.90
(m) Cumene, C_9H_{12}	−1.26	0.04

[a]Reference 19.

Modelling the charge mobility and the J-E curves from trap distributions [2]

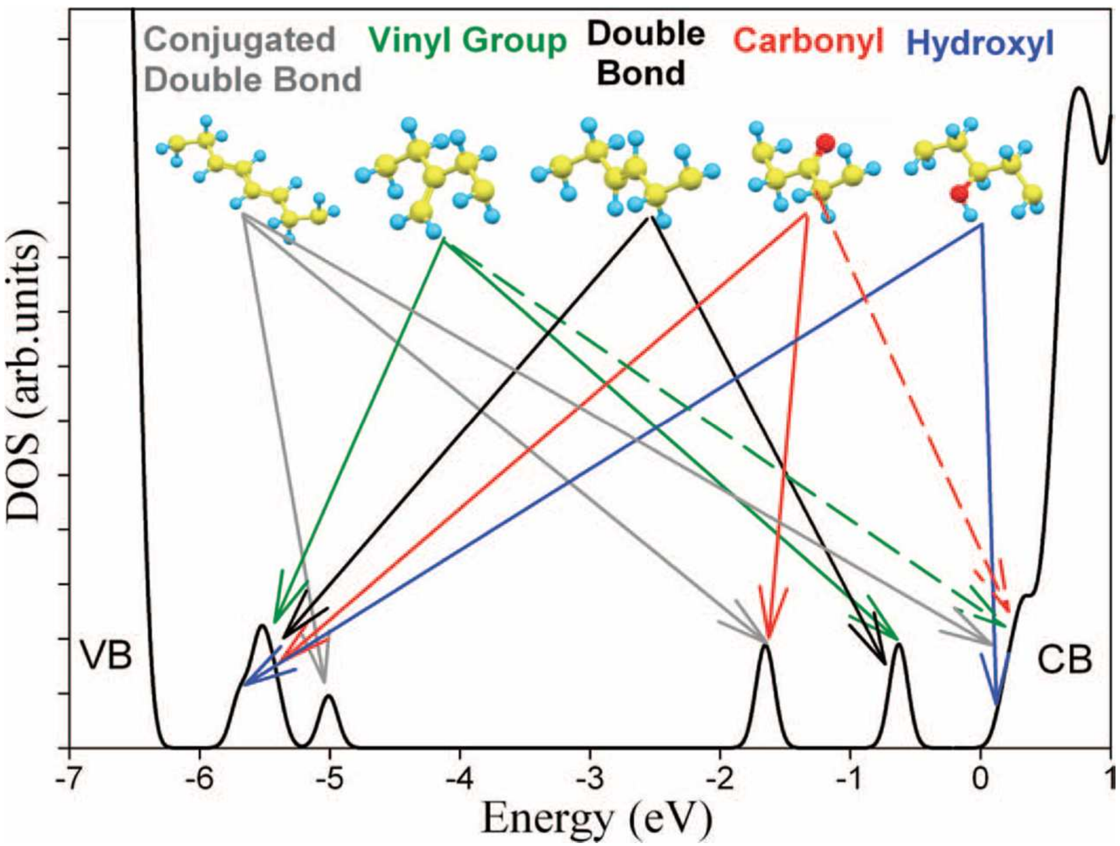


[1] M. Meunier, N. Quirke, and A. Aslanides, Journal of Chem. Phys. **115**, pp. 2876-2881 (2001)

[2] J. A. Anta, G. Marcelli, M. Meunier, and N. Quirke, J. of Appl. Phys. **92**, 1002 (2002)

Examples of results from microscopic models

Hussayyin et al [3]



[AA] A. Huzayyin, S. Boggs, R. Ramprasad, IEEE Electrical Insulation Magazine, 28, pp. 23-29, 2012

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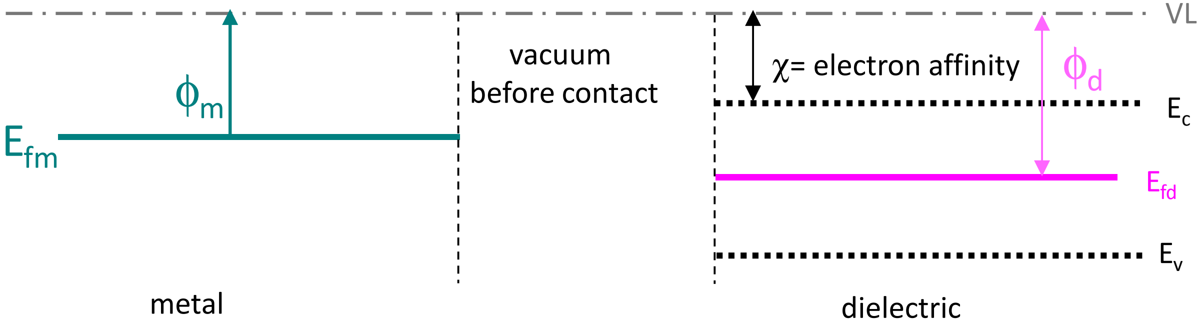
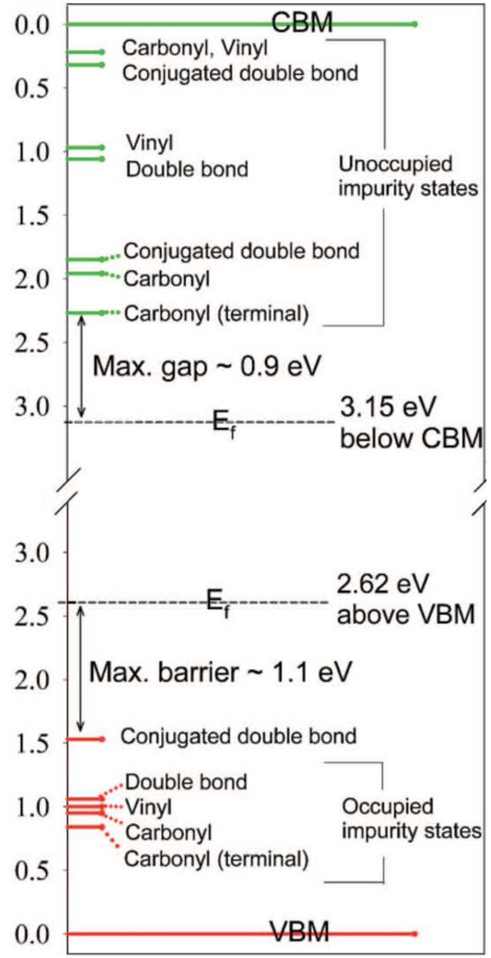
Examples of results from microscopic models

Hussayyin et al [3]

For Pt/PE **interface**

Understanding the metal/PE **interface**





[3] A. Huzayyin, S. Boggs, R. Ramprasad, IEEE Electrical Insulation Magazine, 28, pp. 23-29, 2012

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  - + Density Functional Theory (DFT) calculations – quantuum mechanical computations

- **Macroscopic models:**
  - + conductivity / permittivity as a function of field and temperature
  - + based on experiments

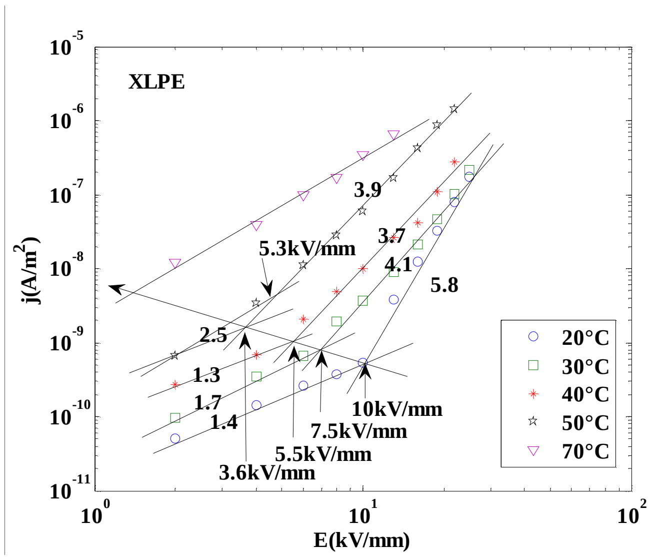
☺ macroscopic characteristics	☹ no prediction of space charge behaviour
☺ description of complex systems	☹ no access to transient mechanisms

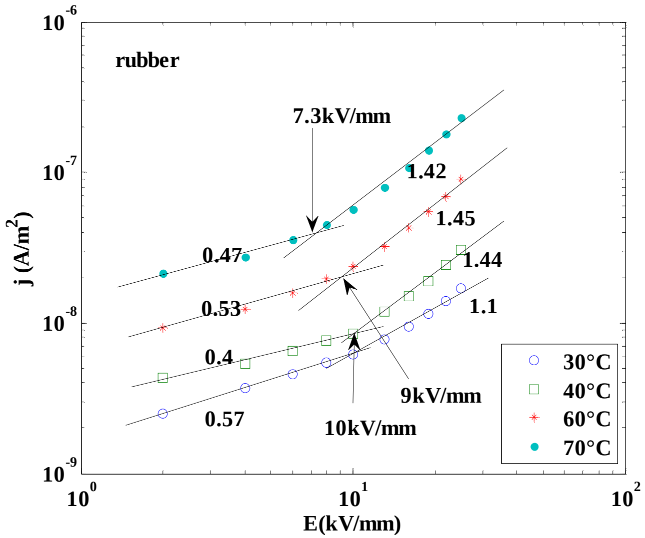
- **Semi-microscopic – Intermediate - models:**
  - + transport phenomena
  - + microscopic phenomena with effective parameters

Examples of results from macroscopic models

Vu et al [4]

1. Current measurements function of field and temperature





[4] T.T.N. Vu, G. Teyssedre, B. Vissouvanadin, S. Le Roy, C. Laurent, M. Mammeri, and I. Denizet, *Proceedings of the International Conference on Solid Dielectrics (ICSD)*, p. 413-416, 2013

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Examples of results from macroscopic models

Vu et al [4]

1. Current measurements function of field and temperature

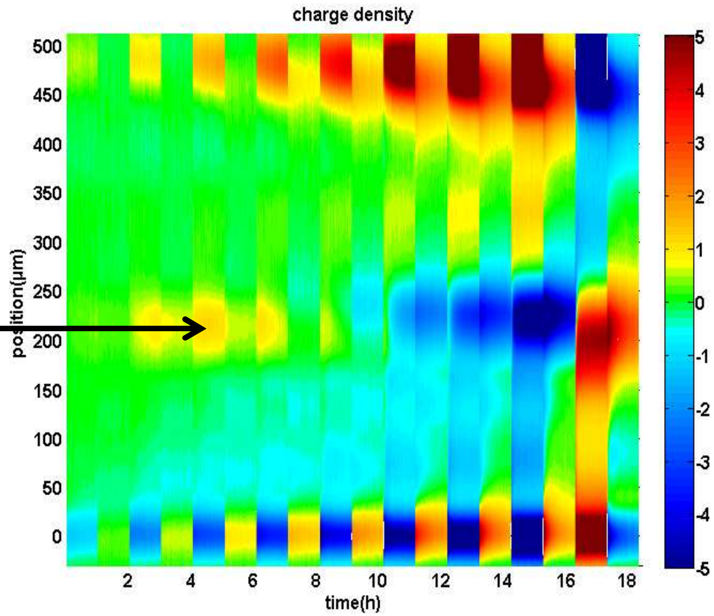
2. Space charge measurements on rubber/PE sandwiches

40° C

rubber

Interfacial charge build up →

XLPE



$$\kappa_{surf}(t) = \int_{x_{i,XLPE}}^{x_{i,MRC}} \rho(t - t_p)dx$$

[4] T.T.N. Vu, G. Teyssedre, B. Vissouvanadin, S. Le Roy, C. Laurent, M. Mammeri, and I. Denizet, *Proceedings of the International Conference on Solid Dielectrics (ICSD)*, p. 413-416, 2013

Examples of results from macroscopic models

Vu et al [4]

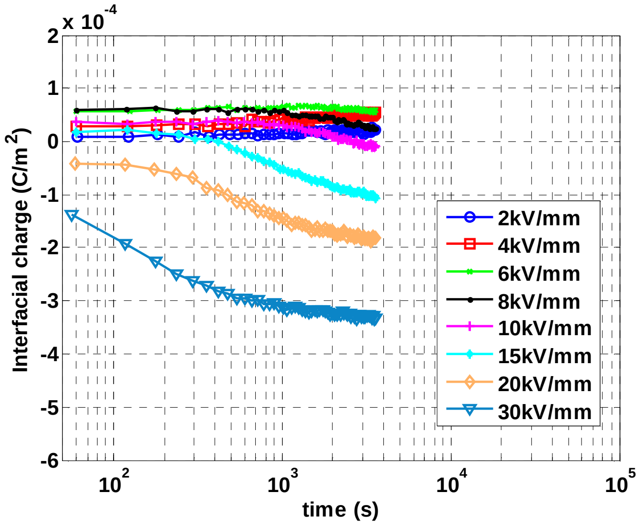
1. Current measurements function of field and temperature

2. Space charge measurements on rubber/PE sandwiches

3. Evaluation of the interface charge

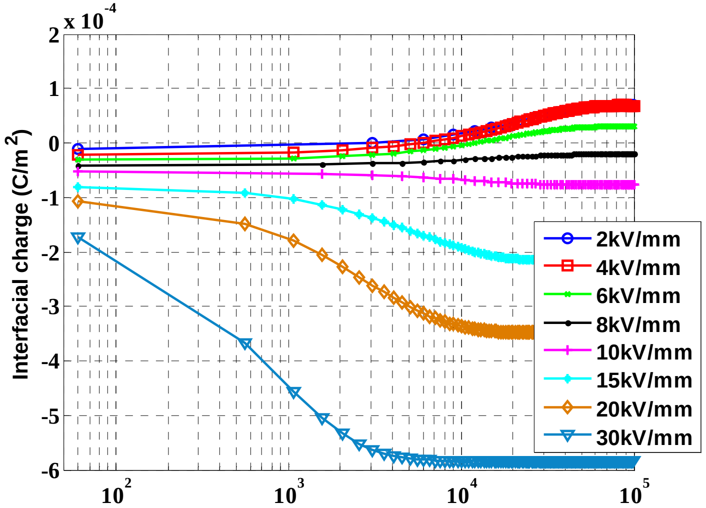
From experiment

$$\kappa_{surf}(t) = \int_{x_{i,XLPE}}^{x_{i,MRC}} \rho(t - t_p)dx$$



From simulation

$$\sigma(T,E) = A.\exp(\frac{-E_a}{k_B T}).\sinh(B(T).E).E^{\alpha}$$



[4] T.T.N. Vu, G. Teyssedre, B. Vissouvanadin, S. Le Roy, C. Laurent, M. Mammeri, and I. Denizet, *Proceedings of the International Conference on Solid Dielectrics (ICSD)*, p. 413-416, 2013

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- **Semi-microscopic – Intermediate - models:**
  + transport phenomena
  + microscopic phenomena with effective parameters

☺ macroscopic characteristics
☺ transient phenomena / space charge ☹ difficult parameterization

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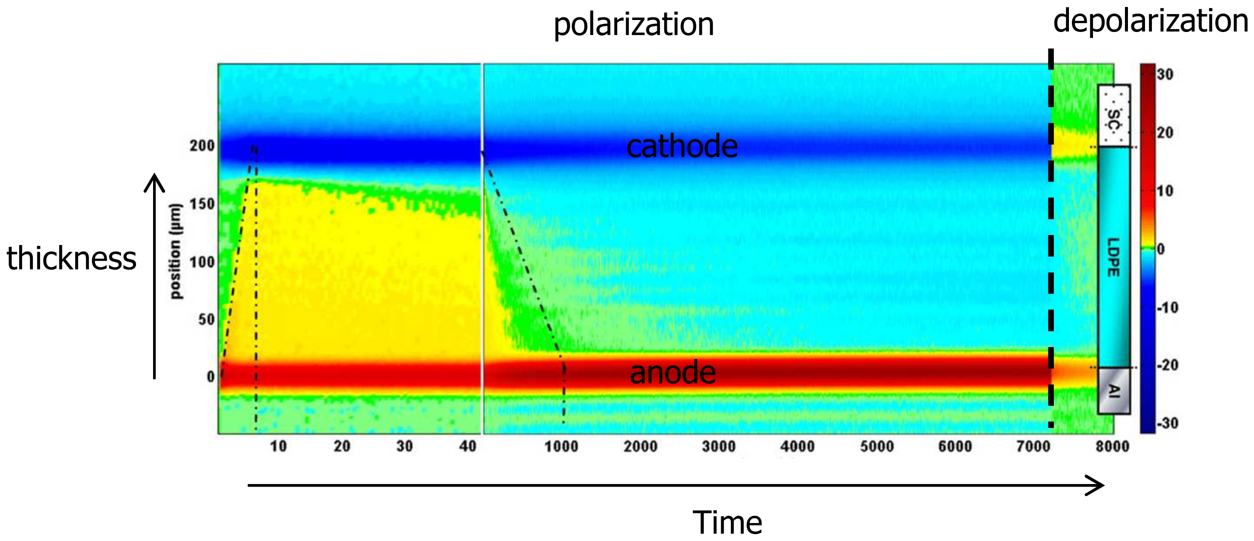
Identification of physical hypotheses

Space charge measurement using the Pulsed Electro-Acoustic method (PEA) [5]

LDPE (low density polyethylene) 200 µm

Applied field -30 kV/mm – polarization 2h – T=40° C



Identification of physical mechanisms:

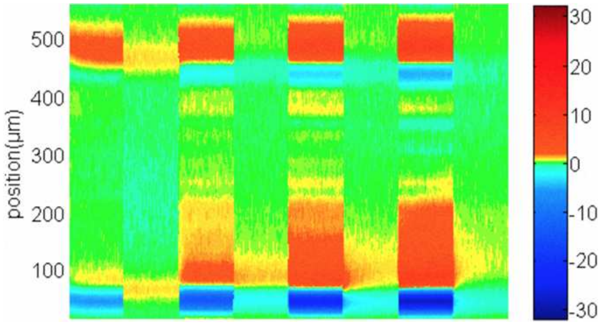
- Injection of positive charges at the anode, negative charges at the cathode
- Transport through the dielectric

[5] Thomas et al, J. Phys. D: Appl. Phys. 44 (2011) 015401

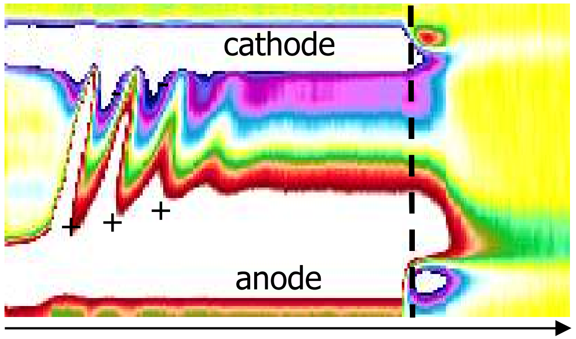
Identification of physical hypotheses

Space charge measurements using the Pulsed Electro-Acoustic method (PEA) [6,7,8]

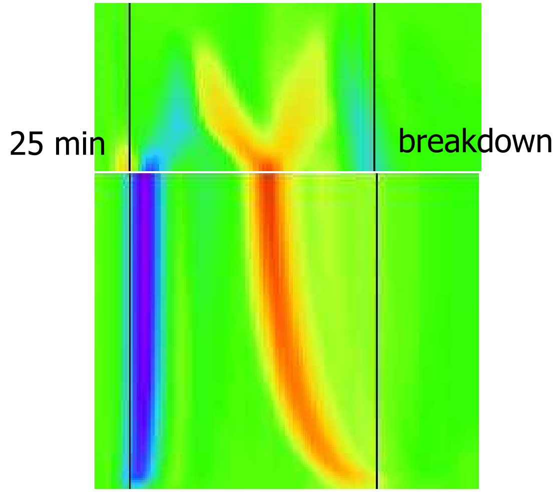
SC/XLPE/SC, thickness 500 µm
degassed 1 day
Polarization/depolarization cycles 2h
Electric field 10, 20, 30, 40 kV/mm



LDPE degassed 2 days, thickness 200 µm
Applied field 80 kV/mm –
polarization 3h – depolarization 3h



LDPE
Applied field
300 kV/mm



Identification of possible physical mechanisms:

o Electronic charge injection and transport ?

o Small transit time -> fast positive charge transport? Or other type of carrier?

o Ionisation? or dissociation?

o Extraction barrier?

o Repetition of the packet initialization at the electrode or surface inhomogeneity ?(initialization of fronts of charge at different point of the surface, integrated with the measurement method)

o …

[6] Vissouvanadin B. et al, ICPADM 2009, Harbin, China, pp 961-964, 2009
[7] Montanari et al, unpublished results
[8] Matsui K. et al, IEEE Trans. Dielectr. Electr. Insul., 12, 406-415, 2005

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Charge generation

Hypothesis: Dielectric sandwiched between two electrodes

➡ injection of charges from electrodes

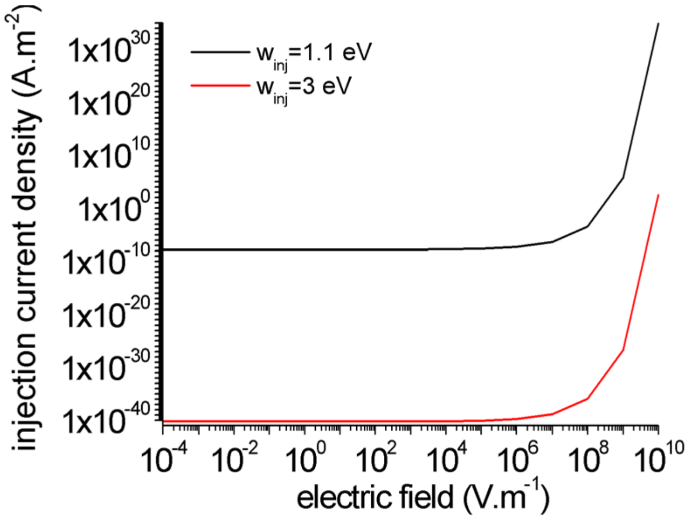
Fowler-Nordheim Emission=
field emission – quantum-mechanical tunnelling

$$j_{F-N} = \frac{e^3 E^2}{8\pi h \phi} \exp\left[-\frac{4}{3} \left(\frac{2m}{\hbar^2} \right)^{\frac{1}{2}} \frac{\phi^{\frac{3}{2}}}{eE} \right]$$

Schottky injection
= thermionic emission

$$j_S = A T^2 \exp\left(-\frac{e\phi_B}{k_B T} \right) \left(\exp\left(\frac{e}{k_B T(t)} \sqrt{\frac{e E_{electrode}}{4\pi \varepsilon_0 \varepsilon_r}} \right) \right)$$

Φ_B barrier height



example interface Au/PE $\Phi_{theoretical} > 4eV$

Impossible to take into account theoretical barrier height values.

For a small or zero electric field, there is always an injection due to the thermal effect.

Charge generation at interfaces

- Schottky equation used with lower values of the injection barrier height (~1 eV)

- Ohm's law also used

- Development of injection 'laws' improving the injection description:
  - ✓ Modified Schottky (no injection when E_a=0)

$$j_S = AT^2 exp\left(-\frac{w_{e,h}}{k_B T}\right)\left(\left(exp\left(\frac{e}{k_B T(t)}\sqrt{\frac{eE_{electrode}}{4\pi\varepsilon_0\varepsilon_r}}\right)\right)-1\right)$$

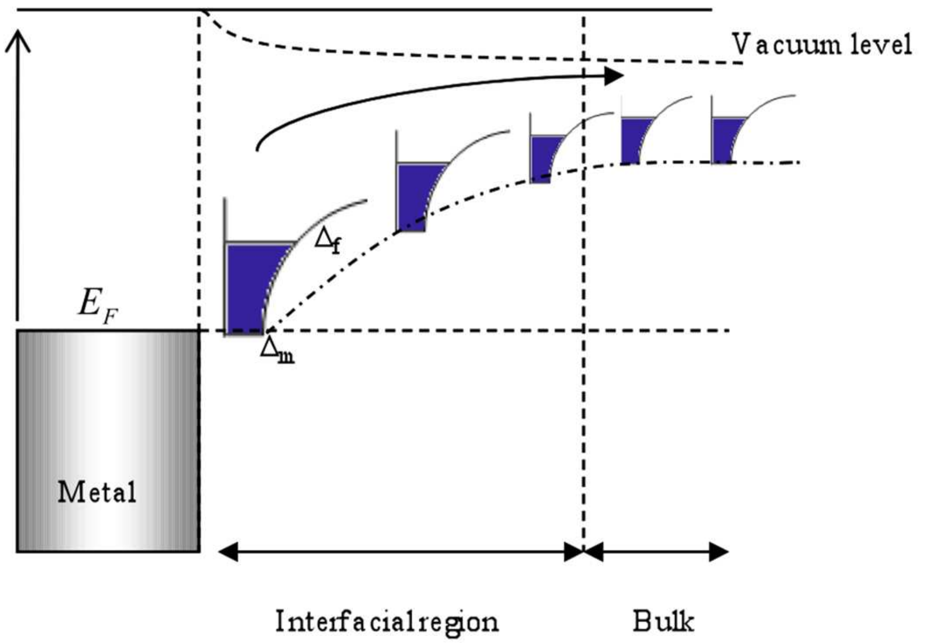
  - ✓ Injection according to a functional approximation for the current density [9]

$$j_{e,h}(x,t) = \alpha \exp(\beta E(x,t))$$

[9] Hare R W, "Modelling space charge in solid dielectrics" *Ph.D. Thesis*, University of Bristol, 1993

Charge generation at interfaces

- o Microscopic description of the interface (higher density of states, deeper traps ..) [10,11]



[10] Junfeng Xia et al, J. Appl. Phys., 109, 034101, 2011
[11] Taleb M et al, IEEE TDEI 2013

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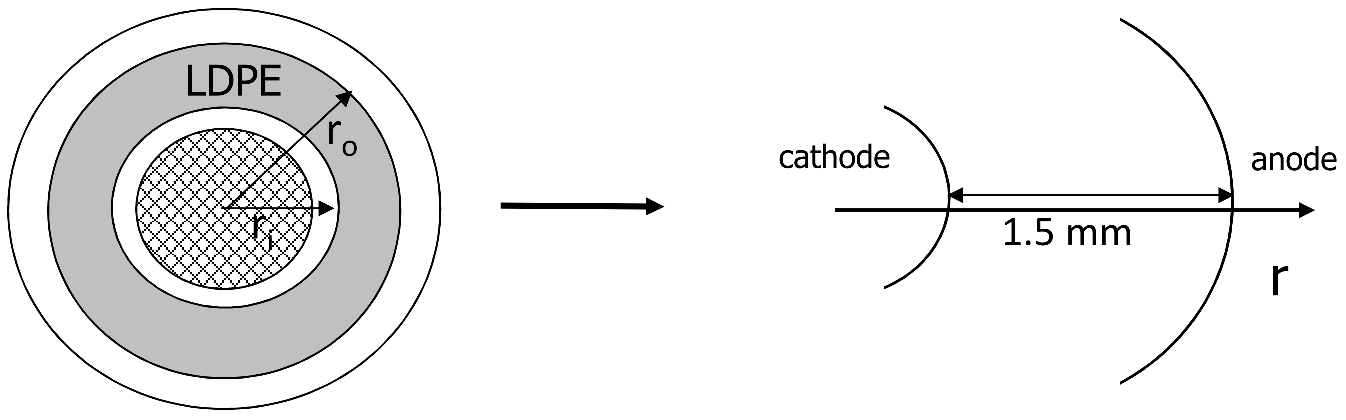
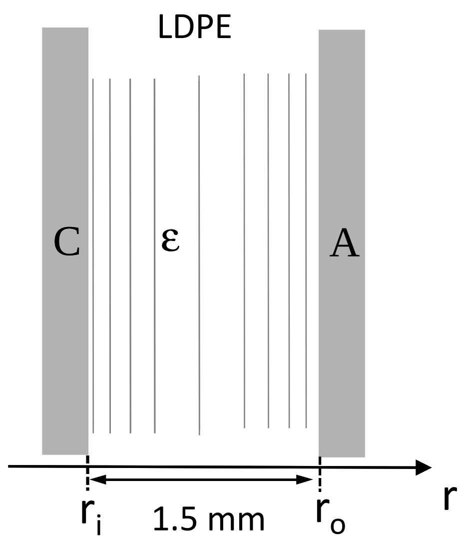
Modelling charge transport in a cable





Experimental and simulation protocol

Mini-cable:

r_i=1,4 mm

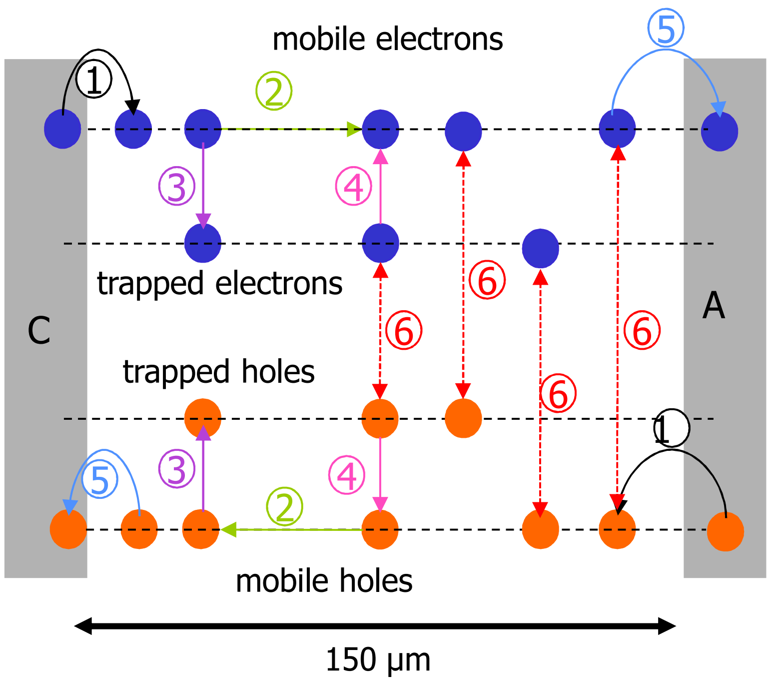
r_o=2,9 mm

d=1,5 mm

V=-30 kV at the cathode

Polarization time: 4h

Physical hypotheses



① **Generation of carrier**
injection at each electrode, modified-Schottky law:

$$j_{e,h} = AT^2 \exp\left(-\frac{w_{e,h}}{k_B T}\right)\left(\exp\left(\frac{e}{k_B T}\sqrt{\frac{eE_{electrode}}{4\pi\varepsilon_0\varepsilon_r}}\right)-1\right)$$

② Transport: hopping mobility taking into account the trapping and detrapping of carriers into shallow traps

③ Trapping: carriers can be trapped into deep traps, using a trapping coefficient B_e and B_h

④ Detrapping: carriers have a detrapping probability function of an activation energy for electrons, for example:

$$D_e = \nu \exp\left(\frac{-W_{detrap}}{k_B T}\right)$$

⑤ Extraction:
the extraction flux of carriers at each electrode is function of the field, for electrons at the anode for example:

$$j_{e,h}(electrode,t) = n_{e,h}\mu_{e,h}E(electrode,t)$$

⑥ Recombination:
Each kind of carrier can recombine with a carrier of opposite polarity, mobile or trapped, using a recombination parameter S_0 to S_3

Models of charge transport: equations to solve

- Poisson equation: $$\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial V(r)}{\partial r}\right)=-\frac{\rho(r,t)}{\varepsilon}$$

ρ : net charge density ($C.m^{-3}$)
ε : permittivity
V : voltage ($V.m^{-1}$)

- Continuity equation: $$\frac{\partial n_a(r,t)}{\partial t}+\frac{1}{r}\frac{\partial j_a(r,t)}{\partial r}=s_a(r,t)$$

n_a : charge density ($C.m^{-3}$)
j_a : flux density ($A.m^{-2}$)
s_a : source term ($C.m^{-3}.s^{-1}$)

- Transport equation: $$j_a(r,t)=n_a(r,t)\mu_a(r,t)E(r,t)$$

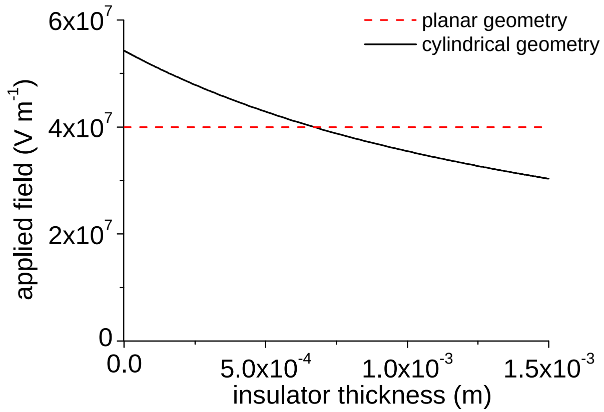
E : electric field ($V.m^{-1}$)
μ : mobility ($m^2.V^{-1}.s^{-1}$)

- Hopping mobility: $$\mu_{e,h}(r,t)=\frac{2\nu d}{E(r,t)}\exp\left(\frac{-ew_{\mu e,\mu h}}{k_B T(r)}\right)\sinh\left(\frac{eE(r,t)d}{2k_B T(r)}\right)$$

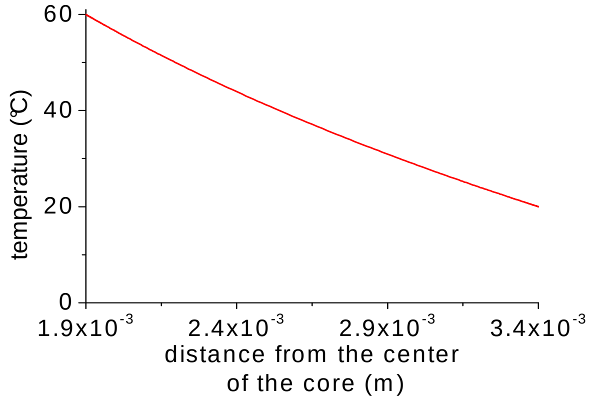
ν : attempt to escape frequency (s^{-1})
d : distance between traps (m)
k_B: Boltzmann constant ($J.K^{-1}$)
$w_{\mu e,\mu h}$: trap depth (eV)

What does a cylindrical geometry imply ?

- Non uniformity of the applied field



- Application of a current in the core of the cable → temperature gradient



$$T(r) = T(r_{in}) - \frac{\ln\left(\frac{r}{r_{in}}\right)\left(T(r_{in}) - T(r_{out})\right)}{\ln\left(\frac{r_{out}}{r_{in}}\right)}$$

Effect on injection

$$j_{e,h} = AT^2 \exp\left(-\frac{w_{e,h}}{k_B T}\right)\left(\exp\left(\frac{e}{k_B T}\sqrt{\frac{eE_{electrode}}{4\pi\varepsilon_0\varepsilon_r}}\right) - 1\right)$$

Effect on mobility

$$\mu_{e,h}(r,t) = \frac{2\nu d}{E(r,t)} \exp\left(\frac{-ew_{\mu e,\mu h}}{k_B T(r)}\right) \sinh\left(\frac{eE(r,t)d}{2k_B T(r)}\right)$$

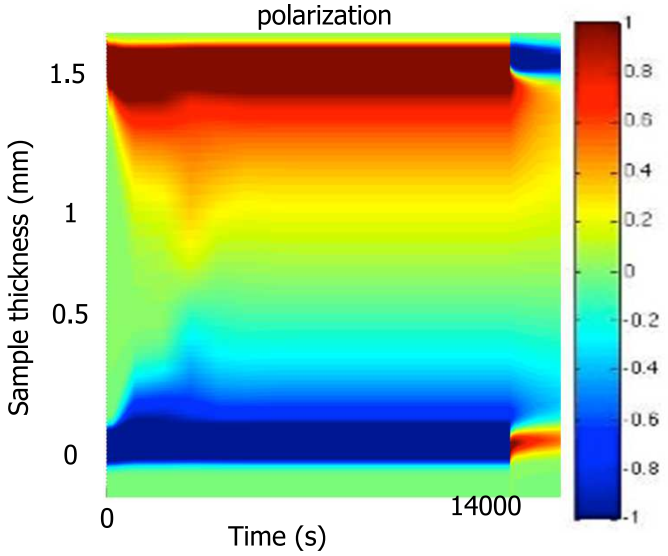
Model parameters

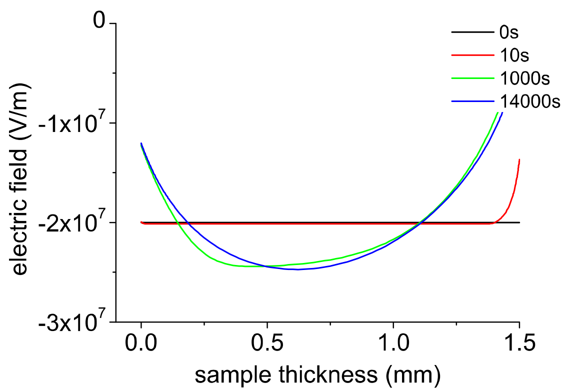
Symbol	value	units
recombination coefficients		
S_0 trapped electron/trapped hole	$4\ 10^{-3}$	$m^3.C^{-1}.s^{-1}$
S_1 mobile electron/trapped hole	$4\ 10^{-3}$	$m^3.C^{-1}.s^{-1}$
S_2 trapped electron/mobile hole	$4\ 10^{-3}$	$m^3.C^{-1}.s^{-1}$
S_3 mobile electron/mobile hole	0	$m^3.C^{-1}.s^{-1}$
trapping coefficients		
B_e electrons	$1.\ 10^{-1}$	s^{-1}
B_h holes	$2.\ 10^{-1}$	s^{-1}
Trap depths (hopping mobility)		
electrons	0.71	eV
holes	0.65	eV
trap densities		
n_{oet} for electrons	100	$C.m^{-3}$
n_{oht} for holes	100	$C.m^{-3}$
injection barrier heights		
w_{ei} for electrons	1.27	eV
w_{hi} for holes	1.16	eV
Detrapping barrier heights		
w_{tre} for electrons	0.96	eV
w_{trh} for holes	0.99	eV

Simulation results: geometry dependence

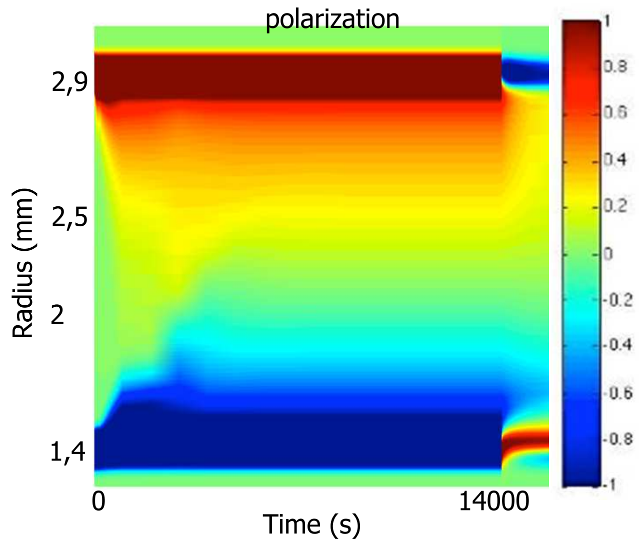
Comparison between planar and cylindrical space charge profiles at T=60° C, V=-30kV

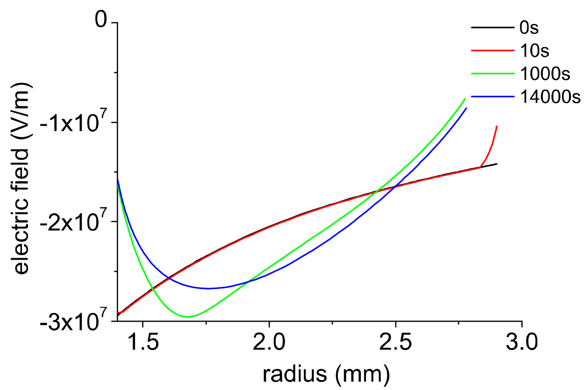
Planar geometry





Cylindrical geometry

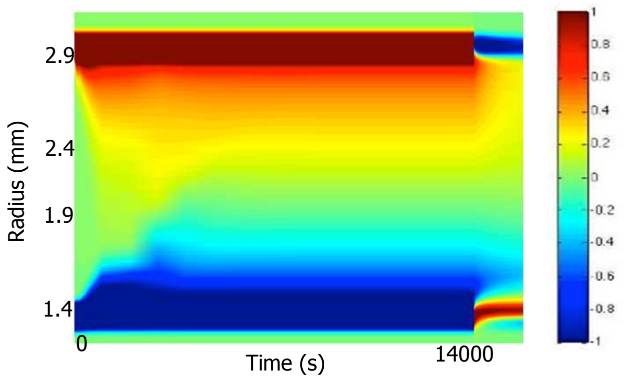


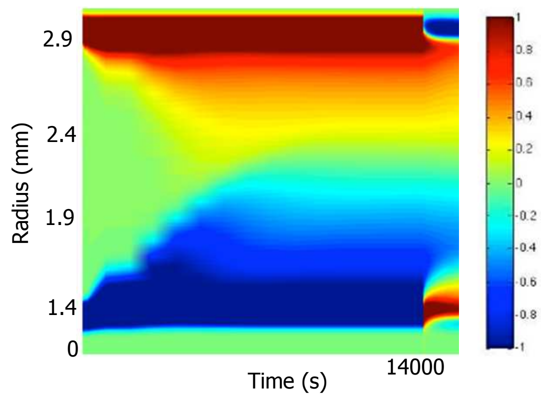


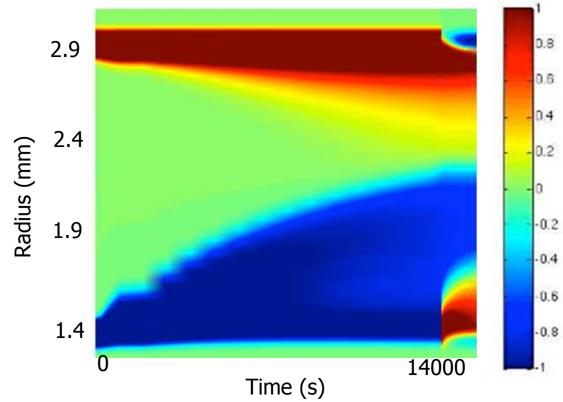
Simulation results: temperature gradients

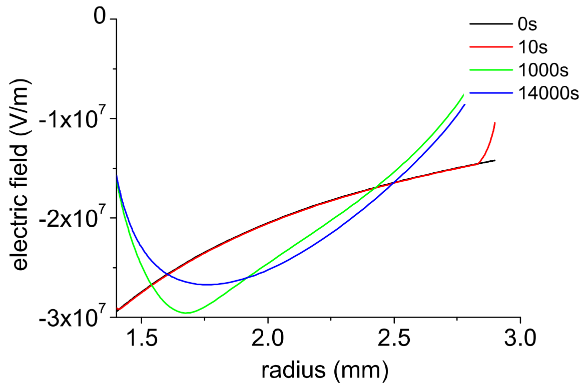
Comparison between cylindrical space charge profiles for V=-30kV, T_{in}=60° C

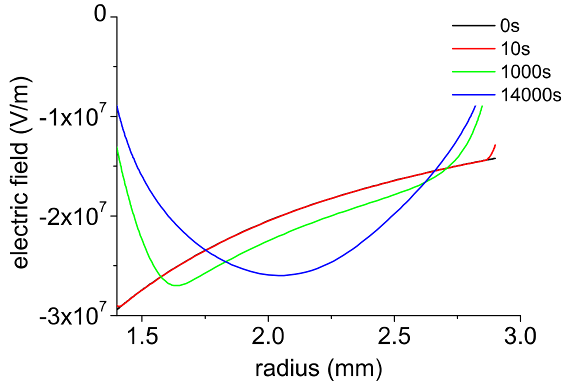
ΔT=0° C	ΔT=10° C	ΔT=20° C

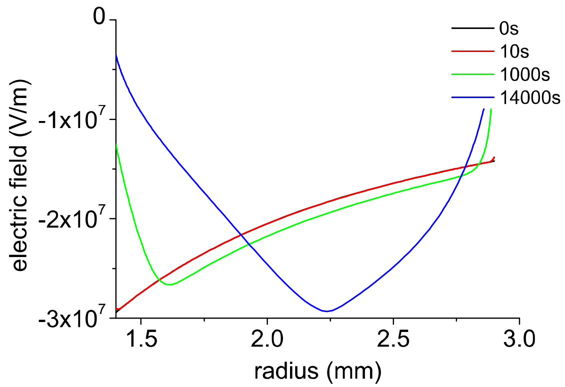












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Conclusions

- Models to help understand the behaviour of a material under electrical stress
- Complementarity of each type of model
- Need to develop the intermediate models with a cylindrical geometry
- Strong effect of temperature gradient on simulated space charge profiles