Numerical modeling of filamentary dielectric barrier discharges in nitrogen taking into account metastable states

Anne Bourdon
Laboratoire EM2C, Ecole Centrale Paris

Yannick Cesses, Pierre Ségur
Laboratoire CPAT, Université Paul Sabatier, Toulouse

Lorentz Center, May 11, 2005
Dielectric barrier discharges at atmospheric pressure

**Homogeneous discharge**

**Advantage:**
Homogeneous discharge → Uniform surface treatment

**Problems:**
High sensitivity to experimental conditions → Transition to filamentary discharge

**Objectives:**
* Identify and understand the physical phenomena which control the homogeneous discharge regime
* Understand the close links between filamentary and homogeneous discharge regimes
Previous works in Pierre Ségur’s team in Toulouse

**Homogeneous discharge**

1990: 1D model in Helium

2000: 1D model in nitrogen
   - Detailed kinetic scheme for nitrogen
   - Current mainly due to $N_4^+$ ions
   - Good agreement between model and experiment (F. Massines, LGET)

**Filamentary discharge**

2001: 2D model in configurations without dielectrics
   - Validated by comparison with previous works (Dhali and Williams, Kulikovsky)

2001: 2D model for a nitrogen DBD discharge
   - Reduced kinetic scheme
   - Simplified transport parameters
   - Photoionization is taken into account
   - Photoemission is the only secondary emission effect

**Present work:**

2D model for a nitrogen DBD discharge
   - Detailed kinetic scheme
   - Accurate transport parameters
   - Influence of different secondary emission processes is considered

Importance of nitrogen metastable states in the gas and on electrodes for secondary emission effects
Filamentary discharge model

- Studied configuration
- Basic equations and numerical schemes
- Transport parameters and source terms

Simulation results of a filamentary discharge propagation

- Discussion on the physical phenomena which control the homogeneous discharge regime
- Further work and open questions
Gas: Nitrogen at atmospheric pressure and 300K
Studied configuration

Gas: Nitrogen at atmospheric pressure and 300K
Applied voltage on the gas at t=0s: 4kV

\[ V_g = V_{\text{anode}} - V_{\text{diel}} \]
Gas: Nitrogen at atmospheric pressure and 300K
Applied voltage on the gas at t=0s: 4kV
Gap distance: 1mm

15% overvoltage at t=0s
Gas: Nitrogen at atmospheric pressure and 300K
Applied voltage on the gas at t=0s: 4kV
Gap distance: 1mm
Dielectrics thickness: 0.65 mm
Dielectrics relative permittivity: 8.6
15% overvoltage at t=0s
Studied configuration

Gas: Nitrogen at atmospheric pressure and 300K
Applied voltage on the gas at t=0s: 4kV
Gap distance: 1mm
Dielectrics thickness: 0.65 mm
Dielectrics relative permittivity: 8.6

Grid: 300 x 200 mesh points
Refined mesh close to symmetry axis and electrodes

Cylindrical geometry
Computational domain radius: 0.6 mm

15% overvoltage at t=0s
Convection-diffusion equation for species i:

\[
\frac{\partial n_i(\vec{r},t)}{\partial t} + \frac{\partial j_i(\vec{r},t)}{\partial \vec{r}} = S_i(\vec{r},t)
\]

with

\[
j_i(\vec{r},t) = n_i(\vec{r},t)\vec{W}_i(\vec{r},t) - D_i(\vec{r},t)\frac{\partial n_i(\vec{r},t)}{\partial \vec{r}}
\]

Poisson’s equation:

\[
\frac{\partial \vec{E}}{\partial \vec{r}} = -\frac{q}{\varepsilon_0} \left(n_+ - n_-\right)
\]
Flux Corrected Transport method (FCT)

High order scheme: 3rd order Quickest scheme (Leonard) extended to variable mesh spacing
Low order scheme: Upwind scheme

Flux limiter: Zalesak limiter

2D treatment: fractional step approach in cartesian coordinates

\[ \frac{\partial n(z, x, t)}{\partial t} + \frac{\partial (w_z(z, x, t) \cdot n(z, x, t))}{\partial z} = 0 \]
\[ \frac{\partial n(z, x, t)}{\partial t} + \frac{\partial (w_z(z, x, t) \cdot n(z, x, t))}{\partial x} = 0 \]

In the next time step, the order is changed

\[ \frac{\partial n(z, x, t)}{\partial t} + \frac{\partial (w_z(z, x, t) \cdot n(z, x, t))}{\partial x} = 0 \]
\[ \frac{\partial n(z, x, t)}{\partial t} + \frac{\partial (w_z(z, x, t) \cdot n(z, x, t))}{\partial z} = 0 \]

=> Used in cylindrical geometry
Numerical solution of Poisson’s equation

Poisson’s equation in cylindrical geometry:

\[
\frac{\partial}{\partial z} \left( \varepsilon(z,r) E_z(z,r,t) \right) + \frac{1}{r} \frac{\partial}{\partial r} \left( r \varepsilon(z,r) E_r(z,r,t) \right) = e\rho(z,r,t)
\]

After integration on a control volume:

\[-W_i V_{i-1,j} - E_i V_{i+1,j} - S_j V_{i,j-1} - N_j V_{i,j+1} + (W_i + E_i + W_j + N_j)V_{i,j} = C_{i,j}\]

where

the potential \(V_{ij}\) is the unknown

\(W_i, E_i, S_j, N_j\) are coefficients depending on position and local relative permittivity

\(C_{ij}\) is a coefficient depending on local net charge density and surface charge density

2D resolution: based on a NAG library subroutine
Nitrogen kinetic scheme

10 species in the model:
\[
\begin{align*}
e^-, N_4^+, N_3^+, N_2^+, N^+, \\
N_2(A^3\Sigma_u^+), N_2(a'^1\Sigma_u^-), N_2(C^3\Pi_u) \\
N, N_2 \text{ (not transported)}
\end{align*}
\]

30 reactions

• Examples of reactions for metastable species:
  Production of secondary electrons in the gas due to metastable species

\[
N_2(A^3\Sigma_u^+) + N_2(a''\Sigma_u^-) \rightarrow N_4^+ + e^- \\
N_2(a''\Sigma_u^-) + N_2(a''\Sigma_u^-) \rightarrow N_4^+ + e^-
\]

Metastable species quenching

\[
N_2(a''\Sigma_u^-) + N_2 \rightarrow N_2(X) + N_2(X)
\]

• Calculation of the \( N_2(C^3\Pi_u) \) radiative state density
  => comparison with experiment
Transport parameters and source terms

Local-field approximation \rightarrow \text{transport parameters and source terms of charged species depend on E/N}

- Electrons:
  
  \begin{align*}
  w(E/N), D_L(E/N), D_T(E/N), \nu_{ion}(E/N), \nu_{exc}(E/N)
  \end{align*}

  \rightarrow \text{Numerical solution of Boltzmann equation for nitrogen}

- Ions:
  \begin{align*}
  w(E/N), D(E/N)
  \end{align*}

- Neutres:
  \begin{align*}
  D
  \end{align*}

Based on experimental data

In previous works, generally simple transport coefficients are taken into account
Transport parameters and source terms

Electron diffusion coefficient

Electron mobility

Ionization coefficient

Transport parameters taken by Kulikovsky in nitrogen
Ion mobilities are based on experimental data for $1 < E/N < 500 \text{Td}$ and extended
• to lower values with Langevin equation
• to higher values with Kawakami analytical expression

Ion diffusion coefficients are based on Einstein relation

Uncertainties on ionization coefficient at high field
Excited species in the model: $N_2(A^3\Sigma_u^+)$, $N_2(a''1 \Sigma_u^-)$, $N_2(C^3\Pi_u)$

Convection-diffusion equation:

$$\frac{\partial n_{exc}(\vec{r},t)}{\partial t} + \frac{\partial}{\partial \vec{r}} \left( D_{exc}(\vec{r},t) \frac{\partial n_{exc}(\vec{r},t)}{\partial \vec{r}} \right) = -\frac{n_{exc}(\vec{r},t)}{\tau_{exc}} + S_{exc}(\vec{r},t)$$

$\tau_{exc}$: radiative relaxation time of the excited species
(36.6ns for the C state, very long for A and a’ states)

$S_{exc}(\vec{r},t)$: production or consumption of the excited state
$\leq$ kinetic scheme

$D_{exc}(\vec{r},t)$: same coefficient for all states based on measurements for the $N_2(A^3\Sigma_u^+)$ state
Secondary emission processes

In a previous work,

- Photoionization in the gas has been taken into account
- Photoemission was the only secondary emission process

In this work,

- Photoionization is neglected (negligible influence in pure nitrogen)
- Photoemission is taken into account
- Secondary emission due to ion and metastable species bombardment is taken into account
Photoemission effect

In the gas, the photon transport equation is:

\[
\frac{\partial \Psi_{ph}(\vec{r}, \vec{c}, t)}{\partial t} + \vec{c} \cdot \frac{\partial \Psi_{ph}(\vec{r}, \vec{c}, t)}{\partial \vec{r}} = \frac{N_{ex}(\vec{r}, t)}{4 \pi \tau_{ex}} - \mu_{ion} \gamma_{ph}(\vec{r}, \vec{c}, t)
\]

\[
\Psi_{ph}(\vec{r}, \vec{c}, t) \quad : \text{the photon distribution function}
\]
\[
\mu_{ion} \quad : \text{absorption coefficient}
\]
\[
\tau_{ex} \quad : \text{radiative relaxation time of N}_2(C^3\Pi_u) \text{ excited state}
\]

The flux incident on the cathode surface located at \(\vec{r}_c\) is:

\[
\vec{\Phi}_{ph}(\vec{r}_c, t) = \int d\Omega \vec{\Omega} \Psi(\vec{r}_c, \vec{\Omega}, t)
\]

The current due to photon impact on the cathode is obtained by:

\[
\vec{I}_{e,c} = ec \sum_{i} \gamma_{ph} \vec{\Phi}_{ph}(\vec{r}_c, t)
\]

Where \(\gamma_{ph}\) is the photoemission coefficient \(\gamma_{ph} = 0.005\)
Photoemission effect

For $\vec{r}_c(0, r)$, the incident flux on the cathode is:

$$
q_{ph,z}^i(0, r, t) = \int_0^L z' dz' \int_0^R r' dr' N_i(z', r', t) \int_0^{2\pi} \frac{d\theta'}{4\pi c \tau_i D^3} \exp(-\mu_{ion} D) \frac{1}{4\pi c \tau_i D^3}
$$

$$
D = |\vec{r} - \vec{r}'| = \left[ z'^2 + r^2 + r'^2 - 2rr' \cos \theta' \right]^{1/2}
$$

If we define:

$$
M(z', r') = \int_0^{2\pi} \frac{d\theta'}{4\pi c \tau_i D^3}
$$

$M(z', r')$ may be pre-calculated and stored

$$
q_{ph,z}^i(0, r, t) = \int_0^L z' dz' \int_0^R r' dr' N_i(z', r', t) M(z', r')
$$

Need to calculate a 2D integral
Electron emission due to ion and metastable species bombardment

\[ \varphi_{Em.Sec} = - \sum_{i=1}^{n_{ions}} \gamma_i \varphi_i - \sum_{j=1}^{n_{metastables}} \gamma_m \varphi_j \]

- \( \gamma \) for a dielectrics may be high:

\( \begin{align*}
\varepsilon_0 & \rightarrow \varepsilon_c \\
\varepsilon_v & \rightarrow \varepsilon_v
\end{align*} \)

Low energy traps (~1-2eV)

Impurity levels

- Order of magnitude

- Ions (value has a small influence): \( \gamma_i = 0.1 \)

- Excited species \( N_2(A^3\Sigma^+_u) \) (6.2eV), \( N_2(a'^1\Sigma^-_u) \) (8.5eV), \( N_2(C^3\Pi_u) \) (11eV)

\( 0.01 \leq \gamma_m \leq 0.5 \) \( \Rightarrow \gamma_m = 0.5 \)
Algorithm

1. Initial conditions
2. Applied voltage
3. Electric field
4. Transport parameters
5. Time step
6. Secondary processes
   - Species transport
   - Source terms
   - Discharge current
7. End

- Geometry
- Initial conditions for particle densities
- Calculation of geometric factors

Resolution of Poisson’s equation using a NAG subroutine
Calculation of mobilities and diffusion coefficients for all particles

FCT method based on a 3rd order QUICKEST scheme and an upwind scheme
4rd order Runge Kutta method
2D evolutions of particle densities, electric field, power and net charge density
Simulation time

Time step in the $10^{-11} - 10^{-13}$ s range

Typical run: 40ns are simulated

2GHz Xeon processor

24h / run
Outline

Filamentary discharge model

Simulation results of a filamentary discharge propagation

First avalanche
Positive streamer
Radial expansion on the dielectrics

Discussion on the physical phenomena which control the Homogeneous discharge regime

Further work and open questions
Gas: Nitrogen at atmospheric pressure and 300K

Applied voltage on the gas at t=0s: 4kV

Gap distance: 1mm

Dielectrics thickness: 0.65 mm

Dielectrics relative permittivity: 8.6

Cylindrical geometry

Computational domain radius: 0.6 mm

Grid: 300 x 200 mesh points

Refined mesh close to symmetry axis and electrodes

15% overvoltage at t=0s
Quasi-neutral plasma spot of gaussian profiles in the r and z directions

$T = 0\text{ns}$

Total number of electrons is $10^5$
First avalanche : 0ns < T < 4ns

![Diagram showing the first avalanche process with reduced electric field and electron movement](image-url)
Time evolution of the electron density

Densité électronique (cm$^3$)

Steps:

* First avalanche

0ns < T < 4ns
Time evolution of the electron density

**Densité électronique (cm⁻³)**

**Anode**

**Cathode**

**STEPS:**

* First avalanche

* Intermediate step

4ns < T < 7ns
Time evolution of the electron density

STEPS:
* First avalanche
* Intermediate step
* Positive streamer
7ns < T < 14ns

Densité électronique (cm$^{-3}$)
Time evolution of the electron density

STEPS:
* First avalanche
* Intermediate step
* Positive streamer
* Cathode sheath

14ns < T < 25ns
Time evolution of the electron density

**STEPS:**

- First avalanche
- Intermediate step
- Positive streamer
- Cathode sheath
- Post-discharge

25ns < T < 40ns
Time evolution of the $N_4^+$ ion density
$N_4^+$ ion density along the dielectrics at the cathode

- Velocity of the discharge radial expansion on dielectrics on the cathode side: $7 \times 10^6 \text{cm/s} \gg$ ion drift velocity for the same electric field ($10^4 \text{cm/s}$)

- Discharge radial expansion on dielectrics is based on a streamer propagation mechanism
Electric field along the dielectrics on the cathode side

- Axial electric field may reach $E/N > 1000 \text{Td}$ at the streamer arrival at the cathode
- Radial electric field remains smaller than axial one
Surface charges on dielectrics

Symmetry axis
Time evolution of the $N_2(A^3\Sigma_u^+)$ density

![Graph showing the density of $N_2(A^3\Sigma_u^+)$ over time with boundaries for cathode and anode.]
Time evolution of the $N_2(C^3\Pi_u)$ density
Discharge current

The different phases of the discharge are clearly shown:

⇒ Avalanche arrival at the anode
⇒ Streamer arrival at the cathode
Temporal evolution of volume averaged species densities

All species first increase in time and decrease slowly
The maximum of species concentrations corresponds to the maximum of current
The decrease of \( \text{N}_2(\text{C}^3\Pi_u) \) is much more rapid
=> in agreement with experiment in similar discharges

In the whole discharge:
• \( \text{N}_4^+ \) is the major ion  \( \rightarrow \) rapid conversion of \( \text{N}_2^+ \) in \( \text{N}_4^+ \)
• High concentrations of excited states
In the post discharge: neutral plasma but high concentrations of charged species
Spatio-temporal evolution of radially integrated $N_2(C^3Π_u)$ density

The different phases of the discharge are clearly shown:
- first avalanche (0-4ns),
- positive streamer (6-16ns),
- cathode sheath formation (16-30ns),
- post-discharge (t>30ns)

The maximum of light correspond to the current maximum (16 to 20 ns)
Secondary emission processes

- During the discharge: the main process is photoemission
- Secondary emission due to ion bombardment remains small during the whole discharge
- When the cathode sheath formation begins, secondary emission due to metastables appears
- During the post discharge, as the electric field decreases, photoemission and ion bombardment decrease, and secondary emission due to metastables becomes the major process
Conclusions about the filamentary discharge propagation in a nitrogen DBD

Three main phases:

- **First avalanche and positive streamer propagation:**
  ⇒ main charged species are created
  ⇒ excited species are created in particular close to the anode

- **Second phase: positive cathode sheath formation (10ns)**
  Major species in the sheath: positive ions
  Sheath depth is about 10 microns, the reduced electric field may reach $E/N > 1000 \text{Td}$
  This sheath is the extension along the dielectrics of the streamer in the gas
  At the beginning its radial extension is the same as the streamer (100 microns)
  and increases with time whereas the maximum of densities decreases

  As the electric field remains sufficiently high during the whole radial expansion,
  excited species in particular metastable states are efficiently produced in the sheath

- **Third phase: post-discharge**
  Mean electric field and mean charged particle energies are close to zero
  ⇒ No more excited species are formed
  ⇒ Metastable species formed during the first two phases have a long life time and
  remain in the gas and close to dielectrics
Discussion on the physical phenomena which control the homogeneous discharge regime

Alternative voltage applied

Numerous filamentary discharges

Metastable species in the gas and close to electrodes accumulate after each voltage alternance

Low field ionization:
- Reactions in the gas:
  \[ N_2(A^3\Sigma_u^+) + N_2(a''\Sigma_u^-) \rightarrow N_4^+ + e^- \]
  \[ N_2(a''\Sigma_u^-) + N_2 \rightarrow N_2(X) + N_2(X) \]
- Secondary emission due to metastables

Previous comparisons with experiments in F. Massines' group =>
Low field ionization is necessary to obtain a homogeneous discharge at Patm

Secondary emission of electrons due to metastable species impact on dielectrics, even if it is a low efficiency phenomenon, could be the fundamental phenomenon explaining the existence of a homogeneous discharge at atmospheric pressure
Further work and open questions

Further work
• Take into account nitrogen vibrational kinetics
• Test the influence of field emission
• Take into account a time dependant voltage
• Test a « real » 2D numerical scheme
• Extend to more complex geometries and mixtures
• Detailed comparison with experiment

Open question
• Problem of charged species in the cathode sheath : high field, small depth
  ⇒ local field approach is no more valid
  ⇒ a microscopic study of electron motion in this region would be necessary
  ⇒ Need for an hybrid model : coupling of a fluid model in regions of low electron energy and a microscopic approach in the sheath