Electrical characteristics of barrier discharge

Tomáš Hoder



Department of Physical Electronics Masaryk University, Brno, Czech Republic



Gas Discharges lecture

Outline

- Historical overview
- Q-V plot (aka Lissajous figure)
- Simplest equivalent circuit of the barrier discharge
- Electrical current in the discharge gap vs. electrical current measured in the external circuit
- Voltage on the gas gap and the electric field parameter
- Application of the electrical analysis for the (not only) spectroscopic plasma investigation
- Understanding the plasma chemistry of low pressure volume and for coplanar barrier discharge in atmospheric pressure air

Barrier discharge - already Siemens had done it...

Wenn man zwei dünne Glas- und Glimmerplatten einseitig mit Stanniol belegt und die nicht belegten Seiten so aufeinander legt, dass ein lufterfüllter Zwischenraum von geringer aber gleichmässiger Dicke sich zwischen ihnen befindet, so erhält man bekanntlich eine Lichterscheinung in dem ganzen lufterfüllten Raume, wenn man den so gebildeten Collector durch eine hinlänglich geladene Leydner Flasche ladet. Diese Lichterscheinung wiederholt sich bei der Entladung des Collectors. Das Leuchten des Luftraums tritt nicht ein, wenn die Flasche sehr schwach geladen ist. Es beginnt bei einer ganz bestimmten Ladung und verstärkt sich von diesem Punkte an mit der Vergrösserung der Ladung der Flasche.

A Dynamical Theory of the Electromagnetic Field. By J. CLERK MAXWELL, F.R.S.

Ozonizea Oxygen

2

W. Siemens 1857 and

U_{Dis}

W. Siemens 1857 Ann. Phys. Chem.

sein. Nimmt man an, dass die Gasmoleelle mit einer Aetherhille un bei der chemischen Verbindung zweier oder

C_D;

C_g -

sein. Nimmt man an, dass die Gasmolecile mit einer solcher Molecile auch eine veränderte Lagerning dethelule und

Seben sind, nehrerer solcher muss bei hillen derselben Wolecule auch eine veränderte Lagernus der Sebindung bie hierdurch bedingte Bewegung der Aeder

hullen derselben Molecule Authertheilchen winstereten. Die lieveränderte Lagerung Ausgangspunkte der Licht- und Wärmewellenzugen ausgeleichen weiden der Jude Wärmewellenzugen beilden weiden der Jude Wärmewellenzugen bilden weiden der Jude Wärmewellenzugen weiden weiden der

Buss 1932, Klemenc 1937, Manley 1943, Samoilovich 1966, Gibalov 1981, Eliasson, Kogelschatz 1983, Heuser 1985, Okazaki 1993, Zhu 1996, Kozlov 2001, Stollenwerk 2007

Manley and his Q-V plots for large scale ozonizers



Manley 1943 Trans. Electrochem. Soc. Kogelschatz 2003 Plasma Chem. Plasma Process.

Simplest equivalent circuit of barrier discharge

... a macroscopic point of view





Liu et al. 2003 J. Phys. D: Appl. Phys. Pipa et al. 2012 Rev. Sci. Instrum.

Kirchhoff's circuit equations and the result



Discharge current correct value?

Tschiersch et al. 2017 J. Phys. D: Appl. Phys. Peeters et al. 2015 Plasma Sources Sci. Technol. Pipa et al. 2012 Rev. Sci. Instrum. Williamson et al. 2006 J.Phys. D: Appl. Phys. $j_R(t) = \left[1 + \frac{C_g}{C_d}\right]i(t) - C_g \frac{\mathrm{d}V(t)}{\mathrm{d}t}$ Merbahi et al. 2004 J. Phys. D: Appl. Phys. Liu et al. 2003 J. Phys. D: Appl. Phys. Bibinov et al. 2001 J. Phys D: Appl. Phys. VS. $I_{\text{discharge}}(t) = I_{\text{meas}}(t) - I_{C_{\text{gas}}} = I_{\text{meas}}(t) - C_{\text{gas}} \frac{\mathrm{d}V_{\text{gas}}(t)}{\mathrm{d}t}$ Reichen et al. 2010 J. Phys. D: Appl. Phys. Massines et al. 2005 Plasma Phys. Control. Fusion Naude et al. 2005 J. Phys. D: Appl. Phys. Bletzinger et al. 2003 J. Phys. D: Appl. Phys. Lomaev et al. 2001 Atmos. Oceanic Optic.

Electrical current balance equation



7

Kulikovsky 1994 J. Phys. D: Appl. Phys. Wang et al. 2006 J. Appl. Phys. Hoder, Bonaventura et al. 2016

Electrical current balance equation + surface charge



Bonaventura, Hoder et al. 2017

Limitations: net charge in streamer head and sheath

2D simulation of the volume barrier discharge in atmospheric pressure air



red arrow denotes the streamer impact onto the cathode creating the conductive channel

Braun et al. 1992 Plasma Sources Sci. Technol.

Limitations: net charge in streamer head and sheath

Correlated current and spatiotemporal development of helium line in barrier discharge plasma jet in atmospheric pressure helium



the streamer impact creating the conductive channel

Sretenović et al. 2014 J. Phys. D: Appl. Phys.

Limitations of current determination



R $j_R(t) = \frac{j_c(t)}{1 - \frac{C_{cell}}{C_i}} \left[i(t) - C_{cell} \frac{\mathrm{d}V(t)}{\mathrm{d}t} \right]$ $U_g(t) = V(t) - \frac{Q(t)}{C_d}$ $E(t) = U_g(t)/g$

red arrow denoting the impact of streamers on the electrodes

Hoder, Synek et al. 2016 Plasma Phys. Control. Fusion

Spectroscopic comparison – helium barrier discharge at 20kPa

Spatiotemporally resolved direct electric field measurement using Stark polarization emission spectroscopy in helium volume barrier discharge





Figure 8. Development of the electric field spatial distribution in DBD in helium at 200 mbar.

Ivkovic et al. 2009 J. Phys. D: Appl. Phys. Liu et al. 2003 J. Phys. D: Appl. Phys.

Spectroscopic comparison – helium at 20kPa

Spatiotemporally resolved direct electric field measurement using Stark polarization emission spectroscopy - comparison



Figure 10. Comparison of calculated cathode fall voltage U_{cal} and measured gap voltage U_g in the DBD at 200 mbar pressure.

Ivkovic et al. 2009 J. Phys. D: Appl. Phys. Liu et al. 2003 J. Phys. D: Appl. Phys.

Spectroscopic comparison – N₂/H₂ mixture at atmospheric pressure in ns-pulsed barrier discharge

Direct electric field measurement in the discharge gap based on coherent anti-Stokes Raman spectroscopy four-wave mixing method



Boehm et al. 2016 Plasma Sources Sci. Technol. Kettlitz et al. 2012 J. Phys. D: Appl. Phys. Pipa et al. 2012 Rev. Sci. Instrum.

derived within the simplest equivalent circuit approach

Spectroscopic comparison – air 30kPa Townsend phase of coplanar barrier discharge prior the breakdown

Effective electric field determined by Townsend alpha coefficient fitting of $\alpha(E/N)$ from high-resolution emission of N₂(C-B) in coplanar barrier discharge



Hoder, Synek et al. 2016 Plasma Phys. Control. Fusion

Pockels effect comparison – helium barrier discharge at atmospheric pressure

Electric field measurement induced by Pockels effect on deposited surface charge



Bogaczyk, Sretenović et al. 2012 Eur. Phys. J. D Liu et al. 2003 J. Phys. D: Appl. Phys.

Determination of capacitances – fully powered large scale reactors (DCSBD, ozonizers, ...)

$$j_R(t) = \frac{1}{1 - \frac{C_{cell}}{C_d}} \left[i(t) - C_{cell} \frac{\mathrm{d}V(t)}{\mathrm{d}t} \right]$$
$$U_g(t) = V(t) - \frac{Q(t)}{C_d}$$
$$q(t) = \frac{C_{cell}}{1 - \frac{C_{cell}}{C_d}} \left[\frac{Q(t)}{C_{cell}} - V(t) \right] + q_0$$

Limited just for full electrode surface coverage by plasma!

E.g. DCSBD at power with full coverage of electrodes by plasma filaments!



Falkenstein et al. 1997 J. Phys. D: appl. Phys. Manley 1943 Trans. Electrochem. Soc. Peeters et al. 2015 Plasma Sources Sci. Technol. Pipa et al. 2012 Rev. Sci. Instrum.

Determination of capacitances – pulsed reactors

Low pressure asymmetric barrier discharge in argon at 100 mbar

Atmospheric pressure symmetric barrier discharge in N₂-O₂ mixture



Applicability of the approach - limitations



$$j_R(t) = \frac{1}{1 - \frac{C_{cell}}{C_d}} \left[i(t) - C_{cell} \frac{\mathrm{d}V(t)}{\mathrm{d}t} \right]$$

$$q(t) = \frac{C_{cell}}{1 - \frac{C_{cell}}{C_d}} \left[\frac{Q(t)}{C_{cell}} - V(t) \right] + q_0$$

$$U_g(t) = V(t) - \frac{Q(t)}{C_d}$$

Limited to barrier discharges which can be described within a single node approximation – i.e. the radial structure is negligible for given spatial- and temporalscale:

- Homogeneous barrier discharges (pulsed or sine applied voltage)
- 2. Nanosecond pulsed barrier discharges
- 3. Spatially confined single-filament barrier discharges
- 4. Multi-filament plasma sources with full electrode coverage

Liu et al. 2003 J. Phys. D: Appl. Phys. Pipa et al. 2012 Rev. Sci. Instrum.

What about not fully powered barrier discharge reactors ... ?

... or middle-sized multifilament discharges without full surface coverage by plasma – what to do?



Peeters et al. 2015 Plasma Sources Sci. Technol.

How to use this? What parameters can be approached...

... besides the cases for mentioned spectroscopy and Pockels effect cases



Effective electric field and power in ns-pulsed single-filament coplanar barrier discharge





Gap voltage, effective reduced electric field and internal discharge current development

22

Instantaneous development of the internal transferred charge, energy and power in the discharge

Compare to Leiweke et al 2013 J. Appl. Phys. Dedrick et al. 2012 Plasma Sources Sci. Technol

Hoder, Šimek et al. 2017

Electron density in ns-pulsed single-filament coplanar barrier discharge

Rough estimate of lower and upper limit of electron density by line-ratio and electrical methods



Other limitation of the method probably reached – plasma channel with high electron density would have less capacitive behaviour as C_g

Hoder, Šimek et al. 2017

23 Compare to Walsh et al 2010 Eur. Phys. J. D

Zhu and Pu 2010 J. Phys. D: Appl. Phys.

Electric field in Townsend phase of coplanar barrier discharge in atmospheric pressure air

Electric field prior the breakdown from Townsend $\alpha(E/N)$ coefficient fitting on N2(C-B) spectra emission

Electric field in the gap from electrical analysis



Electric field at the breakdown instant: 180±30 Td (electrics) and 185±20 Td (fitting) and 200±40 Td from FNS/SPS(E/N)

Hoder, Jánský, Bessiéres et al. 2016

Importance of basic plasma parameters for long-term chemistry (4torr volume BD streamer)



Take-home message

Although spatially unresolved and approximative, the **electrical analysis** according to the simplest equivalent circuit approach **can give important informations about the plasma for (not only) low-density confined plasmas**.

It can gives information about temporal development of the **effective electric field** in the discharge gap, about the **net transferred charge** or **electron density** within the plasma channel or the instantaneous consumed **power in the plasma**.

All these derived informations can **support other methods** applied to investigation of the plasma. For precise analysis an, at least, **2D numerical model for given conditions has to be utilised**.

Single-filament coplanar barrier discharge was studied numerically and experimentally resulting in electric field high-resolution records in quantitative agreement. We plan to compute the generated surface gas chemistry using novel kinetic model of Zdeněk Bonaventura including usage of sensitivity analysis.

Thank you for your attention!



... and thanks to my colleagues and collaborators for the fruitfull discussions and their contribution!

Streamer impact and channel current

2D simulation of the volume barrier discharge in atmospheric pressure air



red arrow denotes the streamer impact onto the cathode creating the conductive channel

9

Braun et al. 1992 Plasma Sources Sci. Technol.

Pre-breakdown phase of pulsed BDs: different pulse widths



>>> new-found local maximum emerging prior to the breakdown of the gap

Hoeft et al. 2014 J.Phys.D:Appl.Phys.

Emission spectra and E/n determination



 \rightarrow access to the electric field determination without any distortion of the discharge – just from its emission

Experimental setup

Without use of theoretical computations we used **single-table setup** including:

- *Corona discharge* as a subject of investigation
- Optical setup with *monochromator*, *photomultipliers* and *TC-SPC module* (resolution of almost 10 ps and 10 μm)
- *Townsend discharge* for electric field calibration



Hoder et al. 2016 Plasma Sources Sci. Technol.

The case of streamer discharge in air

SPS

$$e + N_2(X^1 \Sigma_g^+)_{\upsilon=0} \rightarrow N_2(C^3 \Pi_u)_{\upsilon'=0} + e$$

 $(\Delta E = 11.0 \text{ eV}),$

$$N_2(C^3\Pi_u)_{\nu'=0} \rightarrow N_2(B^3\Pi_g)_{\nu''=0} + h\nu$$

($\lambda_C = 337.1 \text{ nm}$),

$$N_2(C^3\Pi_u)_{\upsilon'=0} + N_2/O_2 \xrightarrow{K_{N_2}^C/K_{O_2}^C} Products,$$

$$\frac{\mathrm{d}n_{\mathrm{C}}(r,t)}{\mathrm{d}t} = k_{\mathrm{C}} \left(\frac{E}{n}\right) n_{\mathrm{N}_{2}} n_{\mathrm{e}}(r,t) - \frac{n_{\mathrm{C}}(r,t)}{\tau_{\mathrm{eff}}^{\mathrm{C}}}$$
$$\frac{1}{\tau_{\mathrm{eff}}^{\mathrm{C}}} = K_{\mathrm{N}_{2}}^{\mathrm{C}} n_{\mathrm{N}_{2}} + K_{\mathrm{O}_{2}}^{\mathrm{C}} n_{\mathrm{O}_{2}} + \frac{1}{\tau_{0}^{\mathrm{C}}}$$
$$= K_{\mathrm{N}_{2}}^{\mathrm{C}} n_{\mathrm{N}_{2}} + K_{\mathrm{O}_{2}}^{\mathrm{C}} n_{\mathrm{O}_{2}} + \sum_{\nu''=0}^{\infty} \frac{1}{\tau_{0\nu''}^{\mathrm{C}}}$$

FNS

$$e + N_2(X^1 \Sigma_g^+)_{\upsilon=0} \rightarrow N_2^+ (B^2 \Sigma_u^+)_{\upsilon'=0} + 2e$$

 $(\Delta E = 18.7 \text{ eV}).$

$$N_{2}^{+}(B^{2}\Sigma_{u}^{+})_{\nu'=0} \rightarrow N_{2}^{+}(X^{2}\Sigma_{u}^{+})_{\nu''=0} + h\nu$$

($\lambda_{B} = 391.5 \text{ nm}$).

$$N_2^+(B^2\Sigma_u^+)_{\upsilon'=0} + N_2/O_2 \xrightarrow{K_{N_2}^B/K_{O_2}^B}$$
 Products.

$$\frac{\mathrm{d}n_{\mathrm{B}}(r,t)}{\mathrm{d}t} = k_{\mathrm{B}} \left(\frac{E}{n}\right) n_{\mathrm{N}_{2}} n_{\mathrm{e}}(r,t) - \frac{n_{\mathrm{B}}(r,t)}{\tau_{\mathrm{eff}}^{\mathrm{B}}}$$
$$\frac{1}{\tau_{\mathrm{eff}}^{\mathrm{B}}} = K_{\mathrm{N}_{2}}^{\mathrm{B}} n_{\mathrm{N}_{2}} + K_{\mathrm{O}_{2}}^{\mathrm{B}} n_{\mathrm{O}_{2}} + \frac{1}{\tau_{0}^{\mathrm{B}}}$$
$$= K_{\mathrm{N}_{2}}^{\mathrm{B}} n_{\mathrm{N}_{2}} + K_{\mathrm{O}_{2}}^{\mathrm{B}} n_{\mathrm{O}_{2}} + \sum_{\nu''=0}^{\infty} \frac{1}{\tau_{0\nu''}^{\mathrm{B}}}$$

Kozlov et al. 2005 J.Phys.D: Appl.Phys

Kinetic scheme, light intensities and ratio



Time-correlated single photon counting technique, cross-correlation spectroscopy



High temporal resolution, but especially high dynamic range and perfect correlation to the studied emission event - the time reference (trigger) is set on the discharge itself!

Ikuta and Kondo 1976 IEE

W. Becker 2005 Advanced time-correlated single-photon counting techniques

LFA - collision frequencies

For justification of **local field approximation** the approach of analysis of the energy-resolved collision frequencies for momentum and energy dissipation as well as the energy-resolved mean free path and energy dissipation length.



Quasi-stationary evolution of the distribution function of the electrons takes place and electrons can be assumed to be in equilibrium with the local field.

Hoder, Loffhagen et al. 2012 Phys. Rev. E

LFA - relaxation of EVDF



EBE and MC results agree well and coincide with the corresponding steadystate electron distribution function components \rightarrow equilibrium values are reached after 10 ps and 5 µm.

Other approach is the study of the electron relaxation in time and space for different reduced electric field strengths E/N:

- ♦ lines represent f_0 , f_1 after **10 ps** of the temporal electron relaxation (solution of the electron Boltzman equation in multi-term approximation).
- \$ symbols denote f₀, f₁ for the 1D spatial relaxation of electrons after a distance of 5 μm using the Monte Carlo method.

Hoder, Loffhagen 2016 PSST



Hoder, Bonaventura et al. 2015 J. Appl. Phys.

$$\nu_{\rm m}(v) = NvQ^{\rm T}(v)$$

$$\lambda_{\rm m}(v) = \frac{1}{NQ^{\rm T}(v)}$$

$$\lambda_{\rm e}(v) = \lambda_{\rm m}(v)\sqrt{\frac{\nu_{\rm m}(v)}{3\nu_{\rm e}(v)}}$$

$$\nu_{\rm e}(v) = Nv\left(2\frac{m}{M}Q^{\rm d}(v) + \sum_{j}Q_{j}^{\rm un}(v)\frac{v_{j}^{\rm un}}{v}\right)$$

Loffhagen 2015