Naučnom veću Instituta za fiziku, Beograd

Pregrevica 118, Zemun

Predmet: izbor u zvanje istraživač- saradnik

Molba

Molim da mi se odobri pokretanje izbora u zvanje istraživač-saradnik. U prilogu dostavljam:

- Biografiju
- Saglasnost rukovodioca projekta
- Potvrdu o upisanim doktorskim studijama
- Spisak i kopije objavljenih naučnih radova

U Beogradu

16.06. 2014. godine

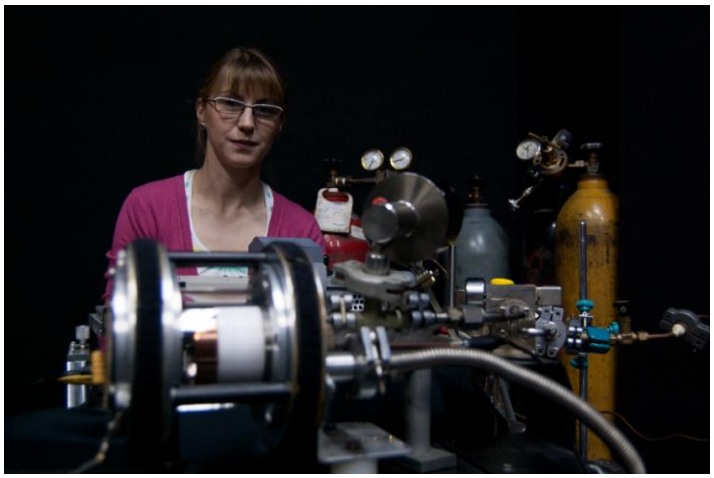
S poštovanjem

Jelena Sivoš

Jelena Sivoš

istraživač- pripravnik

Biografija kandidata



Jelena Sivoš je rođena u Kruševcu 23.01.1983. godine, opština Kruševac, Republika Srbija. Fizički fakultet Univerziteta u Beogradu-smer: Primenjena fizika i informatika završila je 2010. godine. Od 1.1.2011. godine je u stalnom radnom odnosu u Institutu za Fiziku u Beogradu.

Diplomski rad na temu “Proboj i strujno-naponske karakteristike pražnjenja u vodenoj pari" uradila je u Laboratoriji za gasnu elektroniku Instituta za fiziku, pod rukovodstvom prof. dr Zorana Lj. Petrovića i dr Dragane Marić i odbranila ga u decembru 2010. godine na Fizičkom fakultetu u Beogradu. Doktorske studije je upisala sledeće godine na Fizičkom faukultetu Univerziteta u Beogradu – smer: Fizika jonizovanog gasa, plazme i tehnologija plazme.

Ima jedan publikovan rad u istaknutom međunarodnom časopisu, a rezultati njenog rada su prezentovani na više međunarodnih konferencija.

Naučnom savetu
Instituta za fiziku
Pregrevica 118, Zemun

Molim Vas da pokrenete postupak izbora u zvanje *istraživač saradnik* za Jelenu Sivoš, istraživača pripravnika Instituta za fiziku u Beogradu. Kandidat Jelena Sivoš je angažovana na projektu ON171037- Fundamentalni procesi i primene transporta čestica u neravnotežnim plazmama, trapovima i nanostrukturama i III41011 Primene niskotemperaturnih plazmi u biomedicini, zaštiti čovekove okoline i nanotehnologijama. 2011. godine upisala je doktorske studije na Fizičkom fakultetu Univerziteta u Beogradu - smer Fizika jonizovanog gasa, plazme i tehnologija plazme.

Kandidat Jelena Sivoš je, do sada, publikovala jedan rad u međunarodnom časopisu kategorije M_{22}, bila ko-autor na šest predavanja po pozivu na međunarodnim konferencijama, dok su rezultati njenih istraživanja prezentovani i na više međunarodnih konferencija.

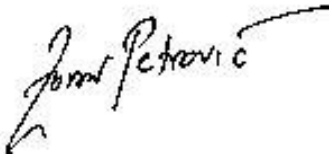
Jelena Sivoš se bavi eksperimantalnim istraživanjem neravnotežnih gasnih pražnjenja, sa posebnim fokusom na proučavanje elementarnih procesa, njihove kinetike i fenomenologije proboja i različitih režima pražnjenja u parama organskih tečnosti. Cilj doktorskog rada Jelene Sivoš je da obezbedi podatke neophodne za razumevanje procesa koji određuju osobine pražnjenja u tečnostima i parama koje su od interesa za razne primene.

Na osnovu svega navedenog, mišljenja sam da je, kandidat Jelena Sivoš, zadovoljila sve formalne i suštinske uslove koji su potrebni za izbor u zvanje *istraživač saradnik*, kako od strane Ministarstva za nauku, tako i od strane Instituta za fiziku u Beogradu.

Predlažem komisiju u sastavu:

1. Dr Dragana Marić, viši naučni saradnik Instituta za Fiziku, Beograd
2. Prof Zoran Lj Petrović, naučni savetnik Instituta za Fiziku, Beograd
3. Dr Nikola Škoro, naučni saradnik Instituta za Fiziku, Beograd
4. Prof Srđan Bukvić, redovni profesor Fizičkog fakulteta Univerziteta u Beogradu

S poštovanjem U Beogradu 16.06.2014.godine



Prof Zoran Lj Petrović

Република Србија
Универзитет у Београду
Физички факултет
Д.Бр.2011/8024
Датум: 13.12.2013. године

На основу члана 161 Закона о општем управном поступку и службене евиденције издаје се

УВЕРЕЊЕ

Сивош (Драган) Јелена, бр. индекса 2011/8024, рођена 23.01.1983. године, Крушевац, Република Србија, уписана школске 2013/2014. године, у статусу: финансирање из буџета; тип студија: докторске академске студије; студијски програм: ДОКТОР НАУКА ФИЗИЧКЕ НАУКЕ.

Према Статуту факултета студије трају (број година): null.
Рок за завршетак студија: у двоструком трајању студија.

Ово се уверење може употребити за регулисање војне обавезе, издавање визе, права на дечији додатак, породичне пензије, инвалидског додатка, добијања здравствене књижице, легитимације за повлашћену вожњу и стипендије.

Овлашћено лице факултета

ФИЗИЧКИ ФАКУЛТЕТ
УНИВЕРЗИТЕТА У БЕОГРАДУ
Број 21172010
Београд, 30. 12. 2010. године

На основу члана 161. Закона о општем управном поступку и члана 4. Правилника о садржају и облику образаца јавних исправа које издају више школе, факултети и универзитети, по захтеву, Сивош (Драган) Јелене издаје се следеће

У В Е Р Е Њ Е

СИВОШ (ДРАГАН) ЈЕЛЕНА, рођена 23. 01. 1983. године у Крушевцу, Србија, уписана школске 2002/2003. године на Студијску групу **ФИЗИКА,** смер: **Примењена физика и информатика,** положила је испите предвиђене наставним планом и програмом наведене Студијске групе и дипломирала на Физичком факултету Универзитета у Београду 29. децембра 2010. године, са средњом оценом 8,34 (осам и 34/100) у току студија и оценом 10 (десет и 00/100) на дипломском испиту и тиме стекла високу стручну спрему (VII степен стручне спреме) и стручни назив

ДИПЛОМИРАНИ ФИЗИЧАР ЗА ПРИМЕЊЕНУ ФИЗИКУ И ИНФОРМАТИКУ

Уверење се издаје на лични захтев, а служи као доказ о завршеној високој стручној спреми (VII степен стручне спреме) до издавања дипломе.

Уверење је ослобођено плаћања таксе.

Д Е К А Н
ФИЗИЧКОГ ФАКУЛТЕТА

Проф. др Љубиша Зековић

Spisak naučnih radova i saopštenja

Naučni radovi M22:

1. Dragana Marić, Marija Savić, Jelena Sivoš, Nikola Škoro, Marija Radmilović-Radjenović, Gordana Malović, and Zoran Lj. Petrović, *"Gas breakdown and secondary electron yields"*, Eur. Phys. J. D vol. 68, issue 6 (2014) 7pp

Saopštenje sa međunarodnog skupa štampano u celini M33:

1. J. Sivoš, N. Škoro, D. Marić, G. Malović and Z. Lj. Petrović, *" Volt-Ampere Characteristics Of Low Pressure Dc Discharges In Water Vapor"*, 26th SPIG (Zrenjanin, Serbia, August 27-31. 2012), ISBN 978-86-7031-242-5 (Published by: University of Novi Sad, Faculty of Sciences Department of Physics, Trg Dositeja Obradovića 3 21000 Novi Sad, Serbia) 273-276

2. V. Stojanović, J. Sivoš, D. Marć, N. Škoro, Z. Lj. Petrović, *" Monte Carlo simulation of electron transport in H_2O vapour"*, 26th SPIG (Zrenjanin, Serbia, August 27-31. 2012), ISBN 978-86-7031-242-5 (Published by: University of Novi Sad, Faculty of Sciences Department of Physics, Trg Dositeja Obradovića 3 21000 Novi Sad, Serbia) 35-38

3. Z.Lj. Petrović, J. Sivoš, K. Karajović, G. Malović and D. Marić, *" Electrical breakdown in water vapor and ethanol"*, 69th Iuvsta Workshop On Oxidation Of Organic Materials By Excited Radicals Created In Nonequilibrium Gaseous Plasma (Crklje na Gorenjeskem, Slovenia, December 9th- December 13th 2012), (Published by: Siovenian Society for Vacuum Technique (DVTS Drustvo za vakuumsko tehniko Siovenije), Teslova 30, SI-l000 Ljubljana, Slovenia) 1-4 **(предавање по позиву)**

4. Z.Lj. Petrović, N. Puač, D. Marić, D. Maletić, K. Spasić, N. Škoro, J. Sivoš, S. Lazović, G. Malović, *"Development of Biomedical Applications of Nonequilibrium Plasmas and Possibilities for Atmospheric Pressure Nanotechnology Applications"*, PROC. 28th International Conference On Microelectronics (MIEL 2012) (Niš, Serbia, 13-16 May, 2012), ISBN 978-1-4673-0237-1 (Published by: Electron Devices Society of the Institute of Electrical and Electronics Engineers, INC.**)** 31-38 **(предавање по позиву)**

5. N. Škoro, J. Sivoš, D. Marić, G. Malović and Z. Lj. Petrović, *" Volt-Ampere Characteristics of Water Vapour Discharges"*, 30th ICPIG (Belfast, Northern Ireland, UK, August 28th – September 2nd 2011), (Published by: Queen's University Belfast, University Road, Belfast BT7 1NN, Northern Ireland, UK) C8-153 (4pp)

6. N. Škoro, D. Marić, J. Sivoš, G. Malović, W. Graham, Z. Lj. Petrović, *"Breakdown and low current discharges in water vapour"*, ECM 112 and 4th ICAPT 2011 (Strunjan, Slovenia, Sep 9th-13th 2011), ISBN 978-961-92989-3-0 (Published by: Siovenian Society for Vacuum Technique (DVTS Drustvo za vakuumsko tehniko Siovenije), Teslova 30, SI-l000 Ljubljana, Slovenia) 164-167

7. Dragana Marić, Jelena Sivoš, Nikola Škoro, Gordana Malović, Thomas Kuschel, Ilija Stefanović, Joerg Winter and Zoran Lj. Petrović, "*Breakdown and Discharge Development in Various Gases and Electrode Configurations"*, 19th Symposium on Application of Plasma Processes Workshop on Ion Mobility Spectrometry (Vrátna, Slovakia, 26th-31st January 2013), (Published by: Department of Experimental Physics, Faculty of Mathematics, Physics and Informatics, Comenius University in Bratislava (Slovakia); Society for Plasma Research and Applications in cooperation with Library and Publishing Centre CU, Bratislava, Slovakia), 33-41 **(предавање по позиву)**

8. D. Marić, J. Sivoš, N. Škoro, G. Malović and Z. Lj. Petrović, *"Low-Pressure Breakdown in Ethanol Vapour"*, 31st International Conference on Phenomena in Ionized Gases (ICPIG) (Granada, Spain, 14th-19th July 2013), Published by: Spanish National Research Council (CSIC); PS4-048 (4pp) (http://www.icpig2013.net/buscador/index.html)

9. J. Sivoš, D. Marić, N. Škoro, G. Malović and Z. Lj. Petrović, *"Breakdown in water vapour and ethanol vapour: heavy particle processes"*, 3rd National Conference on Electronic, Atomic, Molecular and Photonic Physics (Belgrade, Serbia, August 25th, 2013), ISBN 978-86-84539-10-8 (Published by: University of Belgrade, Faculty of Physics, Studentski trg 12, 11000 Belgrade, Serbia) 21-24

Saopštenje sa međunarodnog skupa štampano u izvodu M34:

1. J. Sivoš, N. Škoro, D. Marić, G. Malović and Z. Lj. Petrović, *"Axial emission profiles of Townsend discharge in water vapour"*, ESCAMPIG XXI (Viana do Castelo, Portugal, July 10-14. 2012), ISBN 2-914771-74-6 (Published by: European Physical Society) 9 (2pp)

2. Z. Lj. Petrović, D. Marić, N. Škoro, M. Savić, J. Sivoš, M. Radmilović Rađenović, M. Šuvakov, G. Malović, " *New phenomenology in description of Townsend discharges and gas breakdown: from standard size to micro discharges "*, IC-PLANTS 2011 The 4th International Conference on PLAsma-Nano Technology & Science (Gifu, Japan, March 10-12, 2011), (Published by: Plasma Nanotechnology Research Center) I-07 **(предавање по позиву)**

3. J. Sivoš, N. Škoro, D. Marić, G. Malović and Z. Lj. Petrović, " *Electrical breakdown in low-pressure ethanol vapour* ", 16th International Conference on Plasma Physics and Applications (Magurele-Bucharest, Romania, June 20-25 2013) ISSN 2344 0481 (Published by: INFLPR, Str. Atomistilor, Nr. 409, Magurele, Bucharest, Romania) O18-50 **(предавање по позиву)**

4. J. Sivoš, N Škoro, D Marić, G. Malović and. Z. Lj. Petrović, " *Breakdown in ethanol vapour*", 5th Central, European Symposium on Plasma Chemistry (Balatonalmádi, Hungary, 25-29 August 2013), ISBN 978-615-5270-04-8 (Published by: Research Centre for Natural Sciences, Hungarian Academy of Sciences H-1025 Budapest, Pusztaszeri út 59-67., Wigner Research Centre for Physics, Hungarian Academy of Sciences H1121 Budapest, Konkoly Thege Miklós út 29-33., Diamond Congress Ltd., Conference Secretariat H-1012 Budapest, Vérmezó út 8., HUNGARY), P-FUN6, p.113

5. Zoran Lj Petrović, Jelena Sivoš, Nikola Škoro, Gordana Malović, Dragana Marić, *" DC breakdown in ethanol vapor*", 66th Annual Gaseous Electronics Conference (Princeton, New Jersey, USA, September 30th – October 4th 2013) Bulletin of the American Physical Society, vol. 58, no. 8, HW1.00035 p. 54 (ISSN: 0003-0503 Published by: American Physical Society)

6. Zoran Lj Petrović, Jelena Sivoš, Marija Savić, Nikola Škoro, Marija Radmilović Radenović, Gordana Malović and Dragana Marić, "*New Phenomenology of Gas Breakdown in DC and RF Fields*", VEIT 2013 (Sozopol, BULGARIA, 07 - 11 October 2013) IL-18, pp 37-38 **(предавање по позиву)**

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Topical Review

Gas breakdown and secondary electron yields*

Dragana Marić, Marija Savić, Jelena Sivoš, Nikola Škoro[a], Marija Radmilović-Radjenović, Gordana Malović, and Zoran Lj. Petrović

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Abstract. In this paper we present a systematic study of the gas breakdown potentials. An analysis of the key elementary processes in low-current low-pressure discharges is given, with an aim to illustrate how such discharges are used to determine swarm parameters and how such data may be applied to modeling discharges. Breakdown data obtained in simple parallel-plate geometry are presented for a number of atomic and molecular gases. Ionization coefficients, secondary electron yields and their influence on breakdown are analyzed, with special attention devoted to non-hydrodynamic conditions near cathode.

1 Introduction

It is often said that atomic and molecular collisions define the physics of non-equilibrium (so-called low-temperature) plasma. However, in plasma modeling, where space charge and field profile effects intervene with atomic and molecular collisions, often it is claimed that the collisional cross sections, rate coefficients and swarm transport data do not need to be very accurate as the processes are so complicated that high accuracy is not required. Gas breakdown, on the other hand, is the point where inaccuracies of the atomic collision and swarm data are amplified and at the same time the conditions for the breakdown often define the operating conditions for the plasma. To illustrate this we may give an example that ionization rate enters the breakdown condition in exponent and also that rate is often exponentially dependent on the gas density normalized electric field E/N. The mean energy and the shape of the distribution function that define the rate (together with the cross section for ionization) are on the other hand strongly dependent on all relevant inelastic processes. Breakdown under DC fields and slowly varying AC fields also depends on surface collisions of ions and atoms. Thus, breakdown condition is a very sensitive projection of atomic and molecular collision and swarm transport physics onto the realm of plasma physics.

Gas breakdown has been studied over 100 years and yet many open issues still remain. In DC discharges, the breakdown is usually described by the standard Townsend's theory [1]. Within the past 20 years, with development of experimental and modeling techniques, it became clear that the standard (basic Townsend's theory as depicted in the textbooks) theory of breakdown and low current discharges (the so-called Townsend's regime) requires improvement. Phelps and coworkers [2–5] initiated a comprehensive revision of the theory in all its aspects.

This revision in the lowest current limit (breakdown) included taking into account the contribution of all feedback mechanisms and space-charge effects in breakdown and low current discharges [5]. These authors only covered one gas (argon) with detailed analysis. This is why we felt that a survey of the existing well documented breakdown data would be of value as the basis for further study on the data and elucidation of the issues in use of secondary electron yields in plasma modeling. All of the presented results were obtained in our laboratory and an utmost care has been invested to avoid the usual problems in determining the breakdown data (often depicted as Paschen curves). Those include variable surface conditions, jumping straight into the glow discharge mode, recording the operating conditions for the glow discharge and also the uncertainties that arise from the long statistical delays in initiation of gas discharges.

For many years swarm experiments have represented the primary source of data for gas discharge modeling, which, on the other hand, was based on the transport theory for swarms. With only very few exceptions, the models are based on the hydrodynamic (in equilibrium with the electric field and spatially uniform) transport data. This is however not applicable in most breakdown experiments as the early stages of the breakdown occur before equilibration of the electron swarm. Thus we present also an analysis of electron excitation cross sections and studies of spatial profiles of emission to separate excitation by electrons and fast neutrals [6]. Our results also allow us to determine the width of the non-hydrodynamic region close to the cathode and the effective multiplication as

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well as the approximate determination of the field distribution in dark Townsend discharges. These data all need to be applied to determine the secondary electron yields and in modeling of plasmas.

Over the past two decades determination of the secondary electron yields [6,7] has had renewed interests, for two reasons. First, a systematic survey [5] has been made of all the processes that participate to secondary electron production and it was shown that the basic assumption of Townsend's theory that ions produce the secondary electrons is correct only in a very narrow range of conditions, while photons and gas phase ionization by neutrals contribute to the secondary electron production in a much wider range of E/N. Most importantly, it became possible to model the observed secondary electron yields in the breakdown by using binary collision (beam to surface) data.

It was shown that it is not possible to use directly the binary collision (beam-surface) data for the analysis of gas discharges and low temperature plasmas as those would have specific distributions of all the relevant fluxes that otherwise might be connected through nonlinear relations. The analysis performed for the breakdown (where all fluxes are in linear relation to the initial flux of electrons) proved to be quite robust and still fit most of the data for the glow discharges [8,9]. Nevertheless, it is possible that for some gases or some plasmas, nonlinearities may prevail and the required model may depart from the breakdown model.

In this paper we present the data on breakdown voltages (shown as Paschen curves) for a large number of gases, we show some examples on how these data are coupled with Volt-Ampere (V-A) characteristics, and we proceed to determine secondary electron yields for rare gases (assuming ions to be the primary agent producing secondary electrons) with the inclusion of the effects of equilibration and proper determination of the ionization growth coefficient.

2 Breakdown voltages and Paschen curves

Breakdown is usually represented by a Paschen curve i.e. dependence of the breakdown V_b voltage on the pd (pressure $p \times$ gap d). Parameter pd is a scaling parameter proportional to the number of collisions over a unit distance. In this respect, a typical sharp increase of the breakdown voltage at low pd-s can be explained by the need to compensate for a small number of collisions. On the other hand, at high pd-s, due to a large number of collisions, breakdown voltage is increased in order to enhance energy gain between collisions, when mean free path is getting shorter and the energy gained between two collisions becomes smaller. In the range of the Paschen minimum, production of charges by ionization and secondary electron emission and losses by attachment, diffusion and drift are well balanced.

In Figure 1 Paschen curves for several atomic and molecular gases are presented. Measurements with H_2, SF_6, CF_4, H_2O and C_2H_5OH vapours are taken with the copper cathode, with 1 cm electrode gap and 5.4 cm diameter [10,11]. For all other gases, stainless steel cathode was used in measurements in 2.9 cm gap and 8 cm electrode diameter [6,12]. Some of the data had preliminary presentation in the second edition of the textbook by Lieberman and Lichtenberg [13].

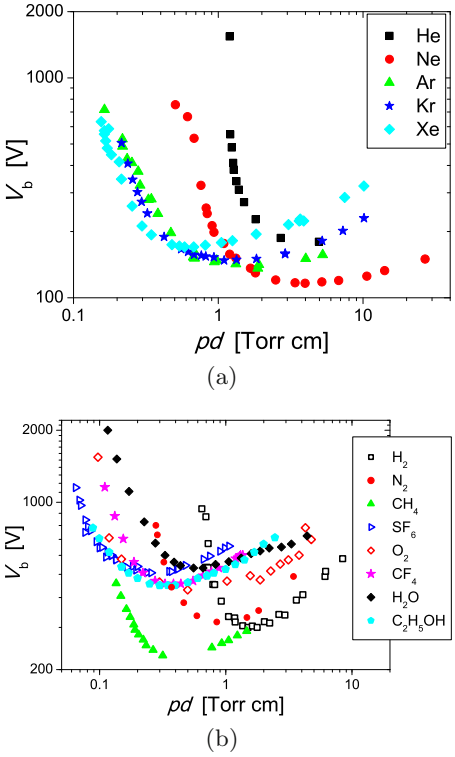

Fig. 1. Paschen curves for (a) atomic gases: Ar, He, Ne, Xe, Kr [6] and (b) molecular gases H_2, SF_6, O_2, CH_4, N_2 [12], CF_4 [10], and H_2O [11] and C_2H_5OH vapours. Measurements with H_2, SF_6, CF_4, H_2O and C_2H_5OH vapours were obtained with copper cathode, for other gases stainless steel cathode was used.

For most of the gases Paschen minimum is situated at pd of the order of 1 Torr cm and breakdown voltages are of the order of several hundred volts. In the case of electronegative gases, it is usually shifted towards smaller pd-s and higher voltages. This can be understood from the point of view of the balance of production and losses of charged particles. In electronegative gases, at low E/N i.e. high pd, attachment becomes important. As a loss mechanism for electrons, it will increase the breakdown voltage and shift the Paschen minimum to lower pressures as an even higher E/N is required to provide sufficient ionization.

There are several issues that one has to be aware of in breakdown measurements. Breakdown voltage depends on the gas mixture through identities of ions and on the cathode material. Even more important than the cathode material is the state of the cathode surface – roughness or possible oxide layers and other impurities deposited on its surface either by exposing the cathode to the laboratory environment or during the discharge operation. Sometimes

the state of the cathode surface has larger influence on the Paschen curve than the material of the cathode itself. For this reason, in our experiments cathode surfaces are treated in low current (∼30 μA) hydrogen discharge prior to the breakdown measurements. This procedure proved to give stable conditions during measurements and reproducible results over large periods of time. Even when basic breakdown voltage varies due to surface conditions, the Paschen curve (and also the V-A characteristics) maintain their shape and so normalization onto the breakdown voltage is a good way to analyze the data [5,14].

Another issue that has to be taken into account in experiments is the regime in which the discharge ignites. Breakdown voltage should not be confused with the operating voltage.The point where the discharge operates is at the crossing of the circuit load-line and the Volt-Ampere characteristics. Quite often, especially with a small series resistance and sufficiently large overvoltages, this is in the regime of a glow discharge, where voltage can be significantly smaller than the breakdown voltage. Actual breakdown voltage, in the sense that is represented by the Paschen law, can only be found by extrapolating Volt-Ampere characteristics to zero current in the dark Townsend discharge mode. An alternative technique is to study the pre-breakdown currents [15,16]. Sometimes it is even necessary to record the spatial profile of the discharge in order to confirm the exponential increase of emission from the cathode all the way to the anode, which is typical for low-current Townsend discharge.

It is important to emphasize that, besides the Paschen curves, Volt-Ampere characteristics are essential in understanding the process of breakdown. These data are needed to establish the electric field/energy dependence of the secondary electron yields and as a consequence the slope of the V-A characteristics in the Townsend regime is defined. The slope of the characteristics is typically negative in the low-current region and it reveals the ion energy dependence of the secondary electron yield and field distortion due to the initial growth of space charge [2,3,17]. In practice, for a full description of the discharge a 3D plot should be constructed [18], such as the one shown in Figure 2, with discharge voltage (V), pressure × electrode gap product (pd) and discharge current (i) presented at the axes.

Low-current limit represents Paschen curve and in this case it is projected onto 1 μA as further changes of voltage at even lower currents would be negligible. Measurements are taken in a parallel-plate electrode system, with 1 cm gap, 5.4 cm electrode diameter and copper cathode. Considerable difference between the glow regime and Townsend regime voltages is clearly seen from the characteristics.

3 Model of the gas breakdown and secondary electron yields

Secondary electron emission is one of the key mechanisms of DC breakdown and operation of discharges. Still, there

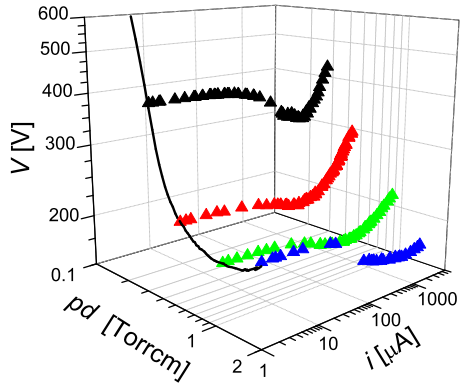

Fig. 2. Experimental V-A-pd characteristics for argon.

is a great confusion in literature in respect to the meaning of the data entering the breakdown condition. In fact, the secondary electron yield data obtained from the gas breakdown have always failed to match the direct measurements in the binary beam-surface experiments. As Phelps and Petrović [5] confirmed in the case of argon, the basic phenomenology of Townsend's theory required extension. Almost constant secondary yield of around 8% for argon ions that has been obtained by ion beams on surfaces cannot be applied to model even the basic low pressure breakdown. While one could justify a greater secondary yield due to additional processes, in the main section of mean energies the yield is actually ten times smaller than that from beam measurements. Phelps and Petrović developed a comprehensive model for argon that included all possible feedback mechanisms – secondary emission by ions, metastables, fast neutrals and photons. They also included back-diffusion of electrons and discussed conditions at the surface where standard gas discharge experiments cannot reach the conditions defined for atomically clean surfaces in ultra-high vacuum. Their study showed that one has to take into account energy dependent yields for each of the species from binary experiments in order to be in accordance with results of direct breakdown measurements. Here, we shall follow the standard procedure to determine secondary yields from the breakdown data and we shall also try to correct some of the problems and provide the data required for such corrections.

Under the conditions of the breakdown and low current Townsend discharges, the effective secondary emission yield (γ) is related to the ionization coefficient (α) in accordance with the Townsends self-sustaining condition:

$$\gamma = \frac{1}{e^{(\alpha/N)\times Nd} - 1} \qquad (1)$$

where N represents the gas number density and d is the gap between the electrodes. $\gamma(E/N)$ may be deduced from Paschen curves by using $\alpha/N(E/N)$ data from the literature [19] as was done in [6]. One may also use an analytic form of $\alpha/N(E/N)$, e.g. Marić et al. [20], as it was shown in [21]. This procedure is the standard one. Perhaps the most important problem in the procedure is that the non-hydrodynamic region close to the cathode (d_0) affects the total multiplication, and therefore the secondary

electron yield obtained from the Paschen curve. The second problem is that the ionization rate taken from the literature may give quite different multiplication as compared with the actual experiment. Even small errors in ionization coefficient result in large discrepancies of the secondary electron yield.

4 Determination of the equilibration distance

It is well-known that hydrodynamic conditions are characterized by transport coefficients that are constant in space and time [22]. However, in low-current electrical discharges at low pressures electrons do not reach the equilibrium state immediately after leaving the cathode. Only at a certain distance from the cathode electrons establish equilibrium with the gas and parameters of electron transport become spatially independent [23,24]. In a simplified approach the width of the non-hydrodynamic region may be used to separate discharge into two regions: one that can be referred to as the non-equilibrium region, with no ionization and the other where ionization behaves as if electrons are in hydrodynamic equilibrium. The problem is then how to determine the delay distance from independent measurements, by using semi-empirical formula such as the one suggested by Phelps and Petrović [5] or by kinetic calculations.

It was shown that inclusion of the effect of equilibration causes a large difference in secondary electron yield data [5], but most authors in the available literature obtain the secondary electron yields from the breakdown data without paying attention to this correction. The role of the equilibration length in determination of the secondary electron yield was studied by Folkard and Haydon [24]. A more detailed discussion of the application of the delay distance and correct determination of the effective electron yield have already been published for the case of argon [6] and for nitrogen [21].

The appropriate form of multiplication factor under Townsend's breakdown conditions is [5]:

$$\gamma = \frac{1}{e^{\alpha(d-d_0)} - 1} \tag{2}$$

where d is the gap between electrodes, and d_0 is the delay distance which has to be passed before electrons reach hydrodynamic equilibrium allowing avalanching characterized by the equilibrium ionization coefficient α. As there is a great need to determine accurate yield coefficients for plasma modeling, there is also a need to establish procedures to determine the equilibration distance.

In our experiments it is possible to obtain equilibration distances from spatial scans of emission. The width of the non-hydrodynamic region d_0 may be used to separate the discharge into two regions. Figure 3 shows two examples of spatial profiles of emission which illustrate the procedure for determination of the equilibration distance and ionization coefficients. In the case of xenon, the non-hydrodynamic width is exhibited as a flat region close to the cathode followed by exponential growth of emission.

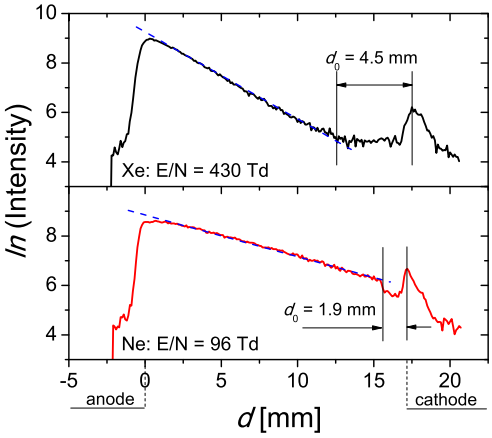

Fig. 3. Examples of the spatial emission profiles in xenon (upper plot) and neon (lower plot), with non-hydrodynamic regions indicated.

In the case of neon, there is even a sudden jump of emission just after the equilibration distance. It is still not clear what is the origin of emission in the region next to the cathode [25], as one would expect that there is no emission in non-equilibrium region. Growth of emission in hydrodynamic region is determined by a single exponential that is in excellent agreement with the equilibrium ionization coefficient [26]. While this is not the most accurate method to determine ionization coefficients, it is useful in some situations when the data are lacking and also to indicate the realistic conditions in a particular system which may be affected strongly by the contamination of the gas. Finally, this is the only direct way to obtain total multiplication as required by the breakdown theory.

When the spatial scans of emission are not available in the experiment that is being analyzed but were available for other experiments, the delay distance d_0 can also be determined by using semi-empirical formulas such as that given in [5] through the expression for the effective value of the electrode potential difference before the exponential growth of the current:

$$V_0 = 16\sqrt{1 + \left(\frac{E/N}{1000}\right)^2}. \tag{3}$$

Probably the best method to produce delay distances is by using Monte Carlo simulations. In this paper we apply a Monte Carlo code that has been well documented in previous publications (details can be found in [27,28]), so only a brief description will be given here. The code is based on generalized null-collision technique [29]. In the code we follow electrons released at the cathode until they reach the anode. The set of cross sections that is used involves inelastic (excitation) processes, ionization and elastic scattering. Each of these processes has associated differential cross sections that are necessary only to establish the angle of scattering. The probability of scattering is determined on the basis of the total cross section. From the simulation of the spatial profile of excitation, one may observe a region next to the cathode where excitation is zero, followed

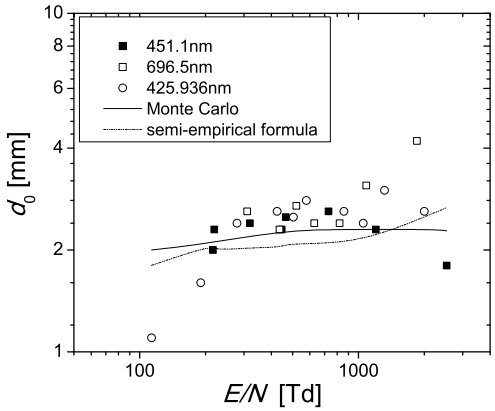

Fig. 4. The dependence of the delay distance d_0 on the reduced field E/N for argon. The delay distances were obtained by three different techniques. Calculations were performed assuming isotropic angular distribution of electrons, the gap between the electrodes of 1.72 cm.

by an exponential growth of emission and finally a growth with the hydrodynamic ionization coefficient. The hydrodynamic region is extrapolated to the zero value and that point determines the distance as applied in equation (2).

In Figure 4 we compare results for the equilibration distance as a function of the reduced field E/N in argon obtained by experiment (symbols), Monte Carlo simulation (solid line) and semi-empirical formula (dashed line). The results obtained by using three different techniques show good agreement, except for the lowest and highest values of the reduced field. It is necessary to consider here the accuracy of experimental determination of the distance d_0 at those values of E/N. At low values of E/N multiplication is very high and it is not so sensitive on the accuracy of determination of d_0 which is small anyway. On the contrary, at high E/N i.e. low pressures, overall multiplication is small, so inclusion of d_0 does not make significant difference. We may say that the agreement between the experimental data, semi-empirical formula and Monte Carlo simulations is excellent for the purpose of determining the secondary yield coefficients. Still, in experiment, due to reflection from the cathode and scatter of light, the results can be significantly scattered, as it is shown in Figure 4, so for the purpose of determination of secondary electron yields, we use results of Monte Carlo simulations when possible.

While Figure 4 shows results for equilibration distance along the Paschen curve, further on, we explore d_0 behavior for the general non-self-sustained conditions. Pressure dependence of d_0 at a fixed $E/N$is shown in Figure 5a and the E/N dependence at a fixed pressure in Figure 5b. In both cases, we present the results obtained using our Monte Carlo simulation code (curve) and semi-empirical formula (symbols). For a fixed reduced field, the delay continuously decreases as the gas number density (pressure) increases. On the other hand, the E/N dependence of the equilibration distance for a fixed gas number density (pressure) shows that the equilibration distance becomes smaller as the reduced field increases (for a fixed gas number density). In both cases, the results obtained by semi-empirical formula and the Monte Carlo simulations are in satisfactorily agreement. The experimental measurements are in fact less reliable than the simulation due to limited spatial resolution and possible scattering of light. Thus we really seek a general agreement and put our confidence in simulations. On the contrary, the measured exponential growth, if defined well and if not overlapping with the contribution of fast neutrals, provides better representation of multiplication in the actual experiment. Agreement between results proves that scaling for the equilibration employed in the development of the semi-empirical formula is appropriate.

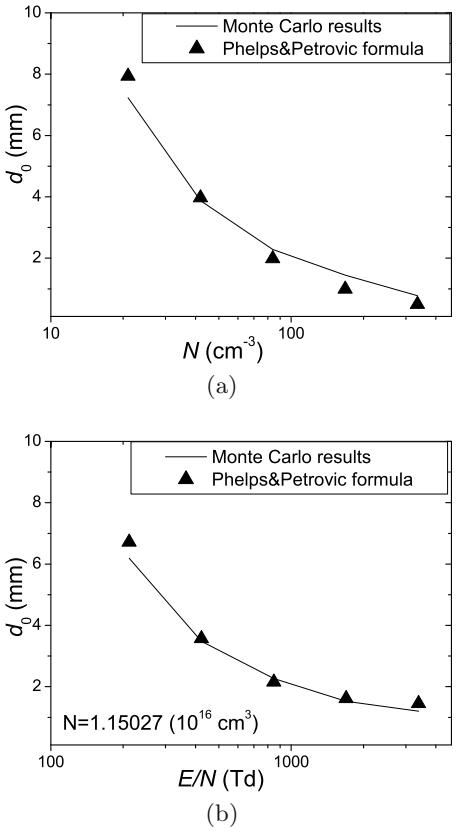

Fig. 5. The dependence of the delay distance on: (a) the gas number density for a fixed reduced field for argon; (b) the reduced field for a fixed gas number density for argon.

In Figure 6 we show calculated equilibration distances for different gasses. We have performed analysis mainly for the rare gases and in a limited sense as compared to Phelps and Petrović [5]. Partly, the reason is that experimental determination of the delay distance in molecular gases is very difficult due to several sources of emission and complex quenching. In those gases we recommend Monte Carlo simulation of the whole system both the delay gap and the exponential growth. In Figure 6 it can be seen that the equilibration distance increases with the atomic mass; however it does not change much for a specific gas in the range of E/N-s investigated here.

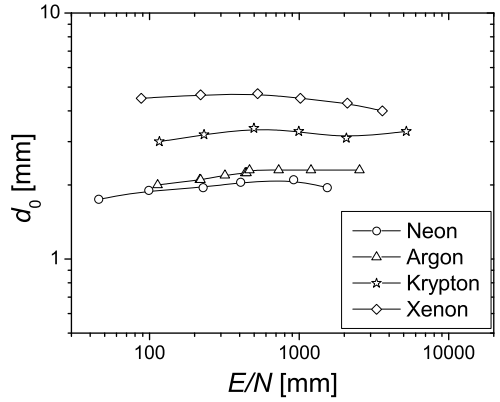

Fig. 6. Equilibration distances for different gases.

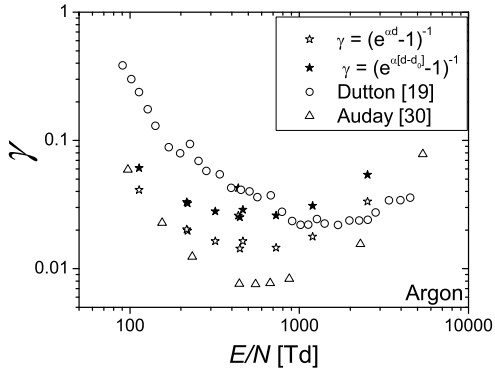

Fig. 7. Comparison of secondary electron yields for argon obtained with different data for ionization coefficients. The first three sets of data were obtained using our Paschen curve, the first two (stars) with our multiplication coefficient without and with the inclusion of the delay distance d_0 in the analysis. The third set (circles) was obtained by using the ionization coefficients from [19] to determine the multiplication. The same ionization coefficients were used in the fourth set [30] but the basis for the results was their measurements of the Paschen curve.

5 Determination of the secondary electron yields and the role of ionization rate

As discussed in previous subsection, the non-hydrodynamic region near the cathode does not necessarily have a significant influence at very low and very high E/N. However, not taking into account the existence of non-equilibrium region can significantly change results for secondary electron yields in medium range of reduced electric fields. In Figure 7 we compare the secondary electron yields in argon obtained by taking into account and not taking into account the equilibration length d_0 (solid and open stars respectively). $\alpha/N(E/N)$ data obtained directly from the experiment are used here to determine γ. As expected, taking equilibration length into account has the largest effect close to the minimum and in the right branch as compared to the left branch. Yet, towards both ends the differences induced by including d_0 diminish. The largest difference between the secondary yields with and without d_0 is a factor of two and it coincides with the minimum of the Paschen curve.

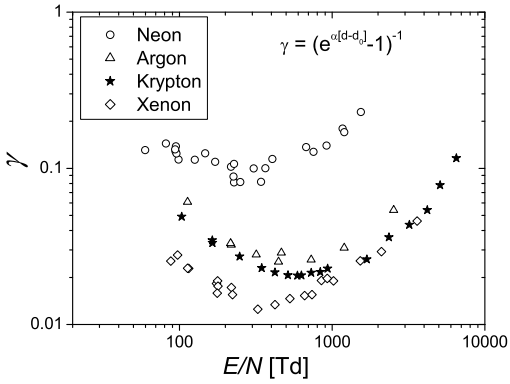

Fig. 8. Secondary electron yields for several different gasses, with the same cathode surface (stainless steel).

Taking the ionization rate from the literature may give a quite different multiplication as compared with the actual experiment and even small errors in the ionization coefficient result in large discrepancies of the secondary electron yield. In Figure 7 we also show secondary electron yields obtained from our Paschen curves by using ionization coefficients from the review [19] which are mostly based on experiments of Kruithiof (circles). These results are up to a factor of 10 different from our data mostly at high E/N.

We also show results of Auday et al. [30] who have analyzed their Paschen curve with the ionization rates from Dutton (triangles). Although those two sets of Paschen curves are apparently quite similar, the differences of yields are considerable, as large as a factor of 10.

For the low values of E/N, γ in our experiment rises more strongly than those obtained by using values of α/N from the literature. This can be explained by the fact that secondary emission of electrons can be due to any combination of numerous mechanisms of varying importance depending on the value of E/N. In the case of small values of E/N, dominant mechanism is the photoelectron emission.

Finally, a similar analysis for the secondary electron yields has been carried out for several other gases. In Figure 8 we show only final results obtained by using the most complete (correct) procedure. As expected the yield increases presumably proportional to potentials of the ion and the metastable states.

6 Conclusions

Measurements of properties of low current discharges which include Paschen curves, Volt-Ampere characteristics and spatial profiles of emission proved to be a fertile basis for modeling of plasmas and discharges. In this paper we gave a short overview of the results of our breakdown studies covering five rare gases and eight molecular gases. We pointed out the most important issues in deducing secondary electron yields from the breakdown and swarm

experiments, compared results obtained by employing different procedures and we presented results for secondary yields for several rare gases obtained by a proper procedure. One should bear in mind that in this analysis the effective coefficients are attached to ion fluxes and a more thorough analysis along the same lines as done by Phelps and Petrović [5] should be performed for all gases together with an analysis of the applicability of the data in higher current discharges.

In conclusion, we may say that the treatment of electron non-equilibrium motion near the cathode includes determination of the delay in reaching the hydrodynamic rates of electron excitation and ionization. The results obtained when the equilibration distance is accounted for allow us to conclude that not taking into account the non-equilibrium region and correct values of ionization coefficients one may make quite large errors in obtaining secondary yields for the relevant particles in the discharge. These differences between the γ coefficients may result in some of the discrepancies between the swarm and the binary collision technique data for γ coefficients, which remains yet to be analyzed.

Monte Carlo simulation provides complete representation of non-equilibrium effect and influence of the electrodes and it is exact representation of breakdown itself, so it should be employed for modeling. A satisfactory agreement between the experimental data and the results obtained using Monte Carlo simulation code and semi-empirical formula proves that our treatment of the electron non-equilibrium behavior close to the cathode is accurate. It also became possible to make more direct comparisons between the secondary electron yields obtained from Paschen's law and from experiments consisting of a beam of ions hitting the surface under high vacuum conditions and separate detailed analyses should be made for all gases that are of interest.

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New phenomenology in description of Townsend discharges and gas breakdown: from standard size to micro discharges

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1 Introduction

In the past fifteen years several issues have opened up in the physics of low current gas discharges. First comprehensive models have been developed that should be able to deal with dc breakdown-low current limit almost exactly [1]. Yet, for the same circumstances, theoretical foundation of the Townsend model provided almost exact representation while giving analytic solutions. It is surprising that there were almost no attempts to compare the two approaches as the latter could be used as a benchmark for the former. Numerical solution to kinetic equations is known to be very accurate provided that the necessary input data are available and Townsend's theory is often employed to provide physical insight. At the same time it was well established that direct application of Townsends theory to analyze the breakdown (Paschen) curves leads to secondary electron yields that may be more than orders of magnitude higher or lower than the data measured directly in binary collision experiments [2].

Resolution of the problem was sought in adding all possible sources of secondary electrons, effects of space charge and non-hydrodynamic processes at or close to the surfaces [2] such as back diffusion, effects of metastables, fast ions and neutrals and photo effect. Such comprehensive approach made it possible to reconcile breakdown and binary collision data for secondary electron emission [2] albeit for the breakdown, i.e. for conditions where all relevant fluxes are linearly proportional to the flux of electrons. Such approach immediately opens a question whether the whole scheme would work for higher current discharges and plasmas where fluxes may be related through complex, non-local and non-linear processes. A limited set of tests [3] provided a relatively good although unexpected agreement of all data.

Another paradigm shift in 100 year old Townsend's theory came from the studies of Volt Ampere (VI) characteristics of discharges. Both VI characteristics and Paschen curve are really needed to determine properly the secondary electron yield [4]. Negative differential resistance [4,5], oscillations [5-7] and constriction [8] are all associated with the development of the space charge [9].

2 Time dependent recordings of the properties of gas breakdown

In addition to new numerical schemes experimental equipment has improved considerably. The use of fast, ICCD cameras allowed recording of temporal development of 2 dimensional pictures thus allowing the anatomy to be used as a guidance to establish the pertinent physical phenomena. Studies of the discharge development in complex albeit stable geometries with somewhat higher current regimes [10] was held in parallel with the measurements for parallel plate discharges [11].

The new results changing the nature of Townsend's theory and phenomenology are related to the variation of the electron cloud during the breakdown and during oscillations, transition to glow and abnormal glow and time dependent coexistence of different modes of discharges.

3 Scaling of the properties of discharges from the standard sizes to micro discharges

Scaling of basic quantities such as pd, E/N and jd^2 scaling where p is the pressure, d is the gap between electrodes, E/N is electric field normalized by the buffer gas particle density and j is the current density, is an important test of compatibility of main physical processes. If we go from d=1 cm (p= 1 Torr) to d= 10 μm (p=1000Torr) the results plotted as a function of pd should be identical. Any breakdown from such behavior may mean that a new physical process has become important [12]. Scaling was found to hold down to d= 10 μm where electric field emission enters the kinetics of secondary electron production [13]. On the other hand this process was often sought for much larger gaps, as an explanation when other factors contributed to the discrepancy between different Paschen curves.

In studies of micro discharges determination of the current density (rather than the actual current) becomes an issue as discharges may happen in narrow channels determined by radial diffusion [14]. Applying basic knowledge of standard size discharges to micro discharges gives basic physical understanding and a number of opportunities to model or diagnose relevant processes in gas discharges.

4 *Kinetic modeling of gas breakdown in RF fields*

Most practical discharges operate in rf and understanding of RF and microwave breakdown is also lacking at the same level as what is known for dc parallel plate discharges. For RF fields one does not need a feedback mechanism such as ion induced secondary electron production as electrons move in both directions and may ionize gas in both if given an opportunity. Current explanations of RF breakdown are based on oversimplified local theories which neglect possible non-hydrodynamic behavior of electrons. We have tried and succeeded in obtaining kinetic Monte Carlo results that give a more complex representation of spatial profiles of ionization and interaction with surfaces. Such profiles help explain the S shaped effective Paschen curves.

5 Conclusion

Several examples presented here show how basic understanding of gas breakdown may be improved by using novel numerical and experimental techniques as well as newly developed theoretical understanding. While the basic phenomena introduced by Townsend still dominate our understanding of breakdown near the Paschen minimum one needs to extend the range of processes that are to be included, change the way those are implemented and look for basic low current limit tests as well as the higher current tests in order to learn how to represent the low temperature plasmas properly

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Effective ionization coefficients in water vapour

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There is a growing interest for electron interaction data in water vapour owing to several fields of application. One way to obtain such data is from low-current discharges and swarm experiments.

We report on measurements of effective ionization coefficients in water vapour discharge for reduced electric fields (E/N) between 600 Td and 5 kTd. Ionization coefficients shown in Fig.1 are determined directly from experimentally recorded emission profiles of low-current dc discharge. At low pressure, in a low current limit of discharge the electric field is assumed to be homogeneous, with typical exponential rise of emission intensity from the cathode towards the anode. Ionization coefficients are obtained directly from the exponential fit of emission profile [1].

In Fig.1 we also show the results from literature [2,3] for comparison. Our results are systematically lower than others. This could be contributed to a different purity of the water sample, or to a different technique of measurements. In [2] authors obtain ionization an attachment coefficient by fitting Townsend theoretical equation for the current growth to the experimentally obtained current growth in steady state Townsend discharge. In [3] ionization coefficients are obtained from measurements of pre-breakdown currents.

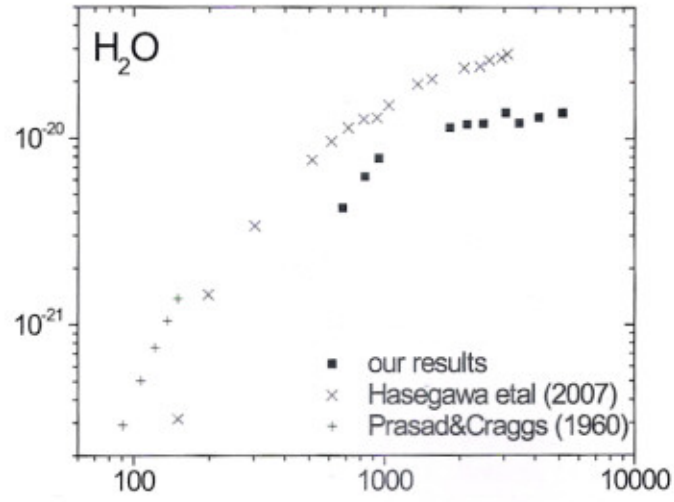

Fig.1. The dependence of reduced ionization coefficient (α/N) on reduced electric field (E/N)

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Volt-Ampere Characteristics of Water Vapour Discharges

N. Škoro, J. Sivoš, D. Marić, G. Malović and Z. Lj. Petrović

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We report on experimental studies of Volt-Ampere characteristics of low-pressure DC discharges in water vapour. Electrical measurements are supported by recordings of axial discharge structure by ICCD camera. Our studies are focused on conditions in the range close to the minimum of the Paschen curve and to the left of it. We analyzed discharge properties for four different electrode gaps (d = 0.55 cm, 1.1 cm, 2.1 cm and 3.1 cm) and for two pressure x electrode gaps (pd = 0.6 Torrcm and 0.3 Torrcm). By comparing results obtained for several electrode gaps we tested the validity of scaling laws for fixed pd value. We also manage to identify dominant processes at different discharge regimes.

1. Introduction

Number of different discharges in vapours and liquids has recently risen up to extent to outline a new distinctive group in the field of non-equilibrium plasmas. While some discharges are successfully applied in plasma devices such as plasma scalpel [1, 2] or miniaturized chemical analyzer [3] many others are still studied in laboratories [4]. All discharges involving water either as liquid electrode(s) or in water-air bubbles systems [5] can be seen as water vapour discharges since working gas contains a significant percentage of water vapour. Furthermore, atmospheric pressure plasmas also operate in an environment with water vapour. In both cases the presence of water vapour influences most of collision and transport properties of the gas [6]. Therefore, studying basic processes governing water vapour discharges and investigation of properties of discharges is of great importance.

This work represents the continuation of a larger study of low-pressure DC discharges in water vapour. Earlier, we reported on breakdown conditions and spatial profiles in low-current limit of the discharge [7]. From measured profiles different dominant processes guiding the low-current discharges were indentified in different parts of the Paschen curve. However, in order to analyze the secondary electron production and to develop realistic models of the discharge it is not sufficient to model just the Paschen curve [8] but also the voltage-current characteristics (V-I) of the discharge [8, 9]. Hence, the investigation of electrical and optical properties of the discharge was extended to higher discharge currents, sweeping though the V-I characteristics. The study also involved changes of pressure and electrode gap in order to obtain measurements for different discharge parameters.

In this paper we show measurements of voltage-current characteristics and spatial profiles for a wide range of discharge conditions. By varying electrode gap and pressure we also investigated the influence of pd (pressure x electrode distance) parameter on spatial properties of discharge.

2. Experimental set–up

Discharge chamber has plane-parallel copper cathode and quartz anode with transparent and conductive platinum film (5.4 cm in diameter) tightly surrounded with quartz cylinder, which prevents the long-path breakdown. The adjustable distance between electrodes is changed between 0.55 and 3.1 cm.

Transparent walls of the chamber allow side-on view and recording of axial discharge profiles. The electrical circuit in the experiment provides highly reproducible and reliable recordings of breakdown voltages and voltage-current (V-I) characteristic of the discharge. The V-I characteristic is recorded by imposing current pulses to a steady working low-current discharge. The pulses are long enough so discharge can reach steady state in higher current regime. In this way gas heating, cathode heating and conditioning are entirely avoided.

High reproducibility of measurements is also achieved by cathode conditioning in a hydrogen discharge prior to the measurements [10, 11] and tested by repeating measurements several times.

Axial discharge profiles are recorded using a sensitive ICCD camera (Andor IStar DH720-18U-03). Time-integrated profiles are recorded in visible spectra and for discharge running in steady state regimes. All emission profiles intensities are scaled relatively.

Water vapour is brought to the discharge chamber from the container through a valve which

allows pressure regulation. From the other side the chamber is connected to a vacuum pump and a pressure gauge. The chamber is pumped down to a high vacuum ($<10^{-6}$ Torr) and then vapour is introduced in the system at a slow flow rate. After the initial period of boiling when dissolved oxygen and other volatile constituents are removed, water in the tube becomes still. Before measurement a moderate pressure of water vapour is maintained in chamber for 1-2 hours in order to achieve saturation of chamber walls.

Room temperature in the vicinity of the discharge chamber was measured and remained constant at around 297K during measurements.

3. Results and Discussion

3.1. Scaling of the *V-I* characteristics

Volt-Ampere characteristics of the water vapour discharge have been measured for four different electrode gaps (0.55 cm, 1.1 cm, 2.1 cm and 3.1 cm) at several *pd* values. We have focused our attention to the *pd*-s that correspond to the minimum of the Paschen curve and to the left-hand branch, as water vapour discharges have not been studied so far under those conditions.

In Figure 1, *V-I* characteristics of the water vapour discharges at different electrode gaps (*d*) and at the same *pd*, are shown. According to our previous measurements of breakdown voltages in water vapour [7], *pd* = 0.6 Torrcm is in the range of the minimum of the Paschen curve. Voltage is shown as a difference between discharge (*V*) and breakdown (V_b) voltage, allowing small variations of the breakdown voltage during the measurements. For the proper comparison of different sets of the data - for the same *pd* and different *d*, one is supposed to use scaling parameter j/p^2 (*j* current density) instead of the current (*i*) in voltage current characteristics. In Fig. 1, we used i/p^2 instead of j/p^2, because effective discharge area hasn't been measured yet for the given conditions [12]. Still, it is worthwhile to comment on the discharge behaviour in this view, at least in Townsend regime, which is radially diffuse, and in abnormal glow regime, which is practically one-dimensional as it occupies entire electrode area [10].

We can distinguish several regions in *V-I* characteristics: low-current Townsend regime, normal and abnormal glow. In most of the cases, we have managed to capture only a very beginning of the abnormal glow discharge mode, due to limiting elements in our electrical circuit (most importantly breakdown voltage of capacitors). There is also a region between Townsend discharge and normal glow, where free-running oscillations occur and it is not possible to obtain steady state values of the voltage and current.

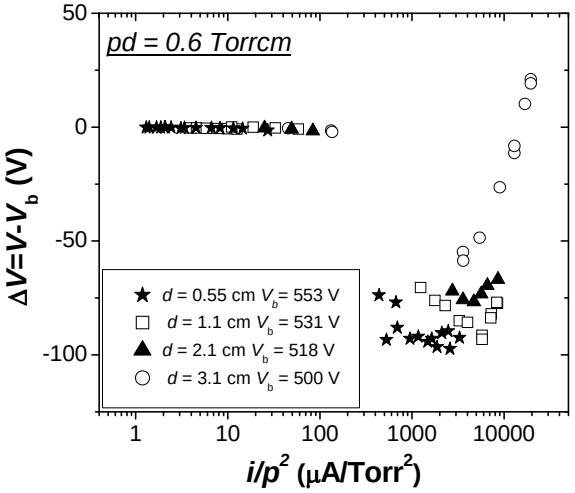

Fig. 1. Volt-Ampere characteristics for low-current discharges in water vapour for *pd* = 0.6 Torrcm and different values of *d*. Different symbols represent different values of the interelectrode distance *d*. *V* is discharge voltage, while V_b denotes the breakdown voltage. Voltage-current characteristics scaled by the parameter i/p^2.

Fig. 1 shows that discharges at different gaps scale well, in the range of low-current Townsend discharge. This indicates that no processes, which would lead to the breakdown of scaling, participate in our discharge (gas or electrode heating, stepwise processes of excitation and ionization, three-body collisions etc.) [4]. Variations in breakdown voltages can be contributed to variations in cathode surface conditions. It is difficult to compare voltages in abnormal glow mode, since we have only limited available data in high current range at shorter gaps. However, it is obvious that voltages are elevated at longer gaps, i.e. lower pressures. Based on our previous studies of the scaling [10], this kind of behaviour could be attributed to additional diffusion losses. When the electrode separation becomes comparable to the diameter of the discharge, radial losses of charged particles become more important. Also, at lower pressures, losses due to a specific electric field distribution around the edges of the cathode can be important [10].

It is interesting to note in Fig. 1 that at the lowest gap concerned here, minimum of the voltage in normal glow is somewhat shifted to lower values of the i/p^2. The discharge is significantly constricted at this pressure, so one would expect that the shift comes from very small effective discharge area. During measurements, we have also noticed that constrictions move over the electrode area, causing

changes in measured voltage and current. Recordings of the radial structure of the discharge, both steady state and time resolved, will be necessary to study more closely the normal glow mode.

3.2. Axial structure of the different regimes of the discharge

Along with voltage and current measurements, we have recorded axial images by the ICCD camera. Establishment of the connection between *V-I* characteristics and the structure of the discharge along the axes, enables us to identify main processes that participate in breakdown and the discharge operation. As an illustration, we show *V-I* characteristics of the discharge at fixed electrode gap, but at different pressures (Fig. 2) with corresponding side-on images of the discharge for typical regimes of non-equilibrium, low-pressure discharges in water vapour (Fig. 3): (a) Townsend discharge; (b) beginning of the normal glow, with distinct radial constriction of the discharge; (c) higher-current normal glow; (d) abnormal glow. We have selected conditions around the minimum of the Paschen curve (0.6 Torrcm) and in the left-hand branch (0.3 Torrcm).

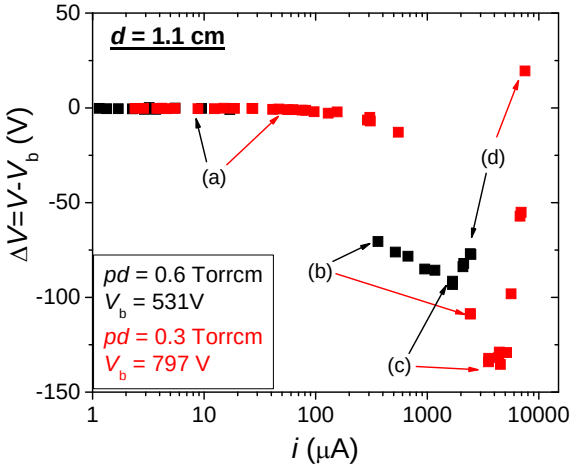

Fig. 2. *V-I* characteristics for two different *pd*-s (0.6 Torrcm and 0.3 Torrcm) at fixed d = 1.1 cm. Labels (a)-(d) indicate different characteristic regimes of the discharge, for which side-on images are presented in Fig. 3.

We can observe standard behaviour of the discharge in both sets of data:
(a) Emission intensity exponentially increases from the cathode to the anode in low-current Townsend discharge.

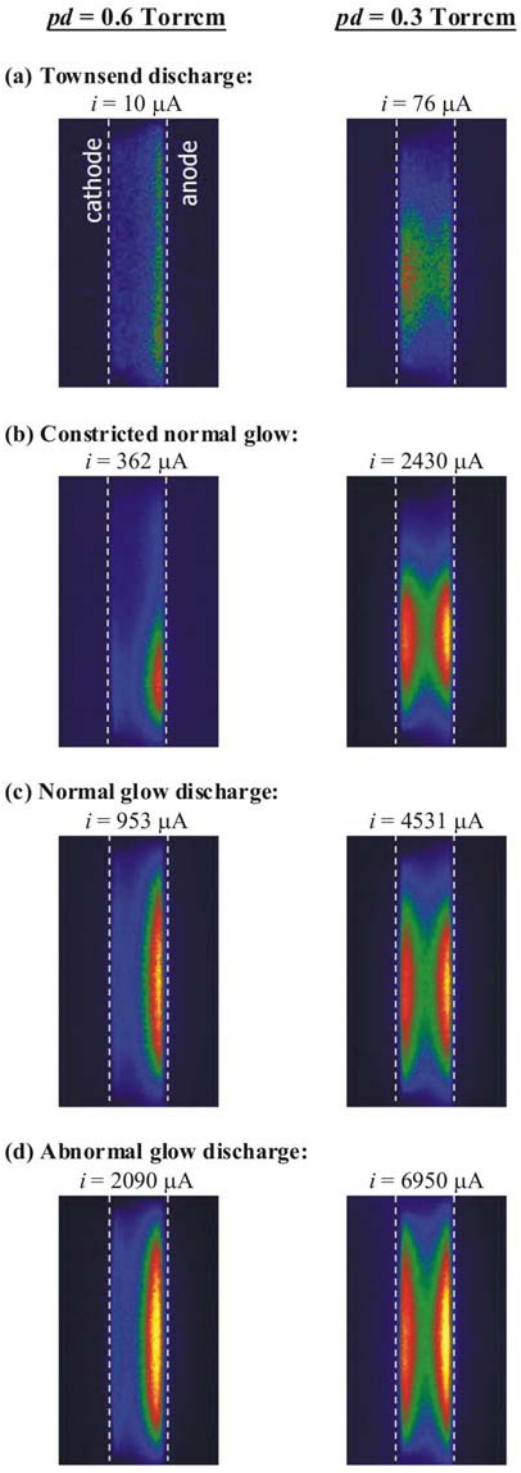

Fig. 3. 2D images of the characteristic regimes of the discharge, taken with ICCD camera in side-on view. White dashed lines indicate position of electrodes. Left-hand column shows the discharge structure at 0.6 Torrcm, while right-hand column is for 0.3 Torrcm. Electrode gap is d = 1.1 cm in both cases. Labels (a)-(d) correspond to labels in Fig. 2.

(b) Distinct constriction of the discharge forms at the beginning of the normal glow, which is followed by the formation of the peak of emission (negative glow), according to formation of the cathode fall.
(c) The discharge is broadening radially with the increase of the current in normal glow. At the same time, peak of emission is moving closer to the cathode. This effect is difficult to observe at such low pressure, as the cathode fall width is close to the electrode distance.
(d) Discharge occupies the entire electrode area in the abnormal glow, and increase of the current can only achieved by increase of the voltage.

All of these features are typical for electron dominated processes of excitation and ionization and space charge effects at higher currents. However, at lower pressures, in the left-hand side of the Paschen curve, when reduced electric field (*E/N*) is high enough, processes of excitation and ionization induced by ions and fast atoms can occur [10,13]. This is revealed through the additional peak of emission near the cathode, which can be seen in Fig. 3 for 0.3 Torrcm through all regimes of the discharge. The most probable candidates for heavy-particles that participate in excitation and ionization are hydrogen ions and fast atoms, as they easily gain energy from the field due to their small mass. Fast atoms are usually created in a charge transfer from fast ions [10,13].

4. Conclusion

We presented results of the measurements of *V-I* characteristics in water vapour for several electrode gaps, supported by imaging of the axial structure of the discharge. Measurements were performed for the conditions close to the Paschen minimum (0.6 Torrcm) and in the left-hand branch of the breakdown curve (0.3 Torrcm). Scaling of the *V-I* characteristics of water vapour discharge at fixed *pd*, for several different electrode distances was analysed. We were able to confirm that scaling holds at 0.6 Torrcm in Townsend discharge. There are some discrepancies in higher-current modes, however, measurements of radial profiles of emission and determination of the actual area of the discharge is required for more detailed analysis.

Furthermore, we discussed influence of *pd* (i.e. influence of *E/N*) to the spatial structure of the discharge. We were able to identify conditions where processes induced by heavy particles (presumably hydrogen fast atoms) become important and even dominant in discharge operation. One of the next steps in our studies will be time-resolved measurements of discharges in water vapour, which will reveal kinetics of basic processes

Acknowledgements

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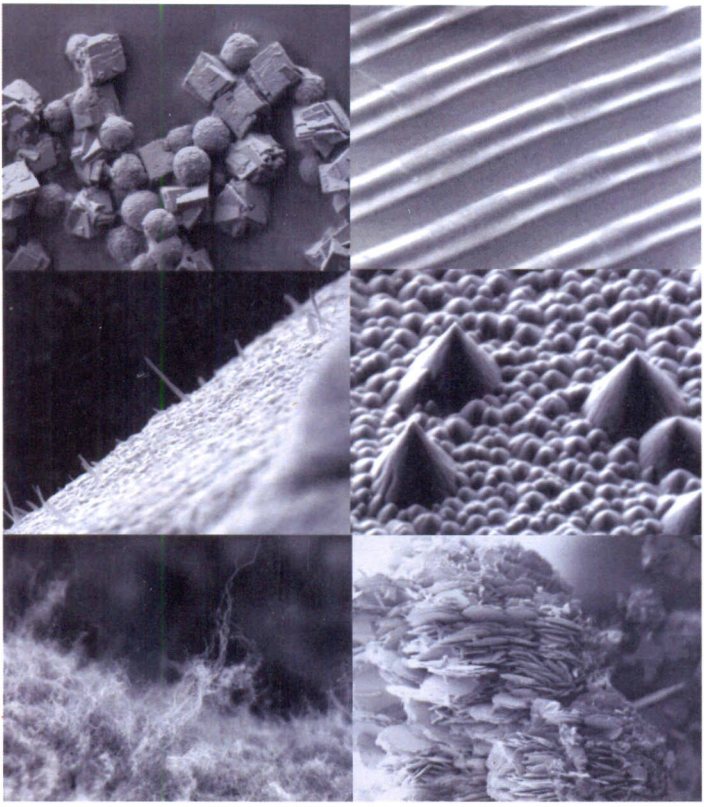

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257634560

Breakdown and low current discharges in water vapour

N. Škoro[1], D. Marić[1], J. Sivoš[1], G. Malović[1], W. Graham[2] and Z. Lj. Petrović[1]

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Wide range of possible applications of non-equilibrium discharges in vapours and liquids drew attention to these discharges [1]. In several cases, investigation resulted in fully developed products, like plasma scalpel [2] or small chemical analyzer [3]. Moreover, discharges involving water either as liquid electrode(s) or in water-air bubbles systems as well as atmospheric pressure plasmas that also operate in an environment with water vapour can be seen as water vapour discharges [4]. Therefore, studying basic processes governing water vapour discharges and investigation of properties of discharges is of considerable interest.

In this paper we show several results of larger systematical measurements of dc breakdown (i.e. Paschen curves) and voltage-current characteristics with spatial discharge profiles. By varying electrode gap and pressure experimental results for a wide range of discharge conditions were obtained.

Discharge chamber has plane-parallel copper cathode and quartz anode with transparent and conductive platinum film (5.4 cm in diameter) tightly surrounded with quartz cylinder. Electrical circuit of the experiment allows us to maintain a stable low current dc discharge while imposing a current pulse at the same time. Axial discharge profiles are recorded using a sensitive ICCD camera (Andor IStar DH720-18U-03) [5]. Water vapour was obtained by evaporation of bi-distilled and deionised water sample at low pressure.

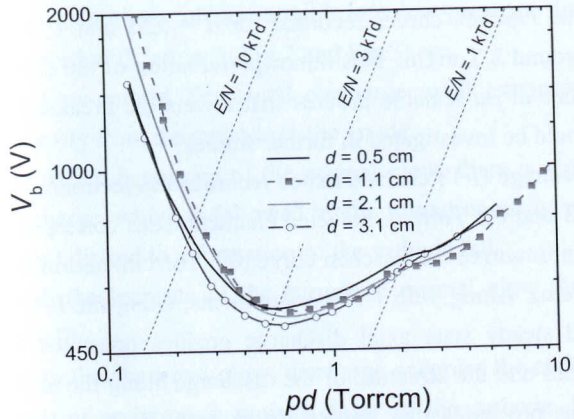

Fig.1 Paschen curves obtained for different electrode gaps (d) in water vapour. The characteristic E/N values are shown on the plot.

Paschen curves, measured for different electrode gaps are presented in Fig.1. All the curves agree well within the experimental uncertainties on the left hand side and around the minimum.

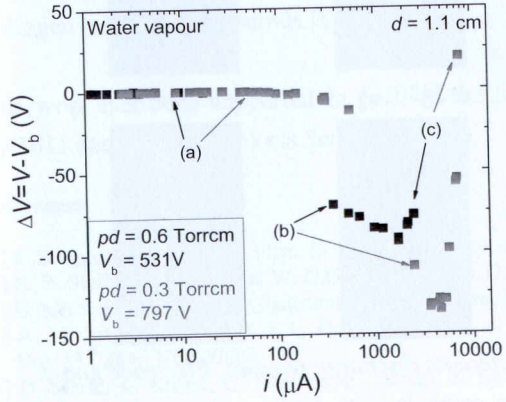

Fig.2 Current-voltage characteristics at d=1.1cm and pd = 0.3 and 0.6 Torrcm. Labels correspond to discharge profiles in Fig.3

On the right hand side, the Paschen curves recorded for d = 0.55 and 1.1 cm have an inflection point around 2 Torrcm. This minor perturbation of the curve appears to be a consequence of pd scalable process that lowers the breakdown voltage. The inflection should be investigated in further studies.

In fig.2 we show current-voltage (I-V) characteristics recorded at electrode gap d=1.1 cm for two pd-s: 0.3 and 0.6 Torrcm. Selected characteristics correspond to conditions around the minimum of the Paschen curve (0.6 Torrcm) and in the left-hand branch (0.3 Torrcm). Along with I-V measurements, using the ICCD camera we have recorded steady state axial discharge profiles presented in Fig.3. The I-V characteristics and the structure of the discharge along the axes, allows us to identify main processes that participate in breakdown and the discharge operation.

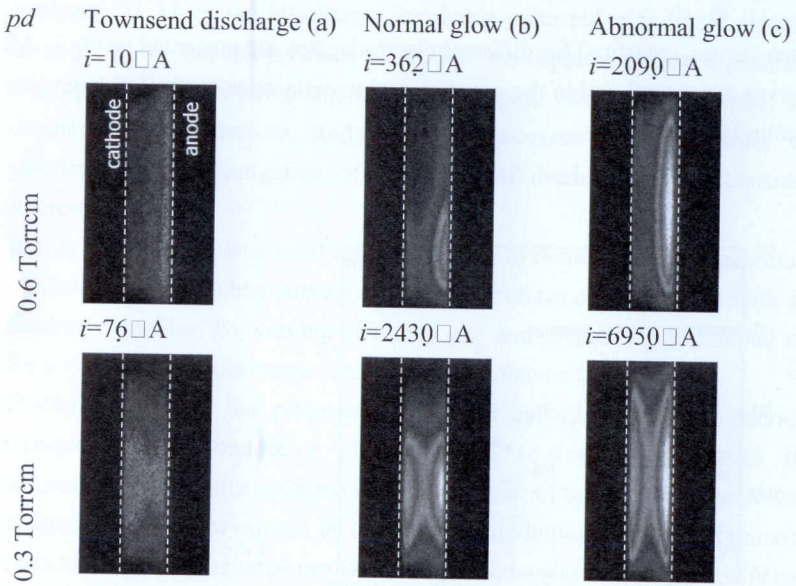

Fig.3 Side-on images of characteristic discharge regimes. Electrode gap is d = 1.1 cm. Brighter colours coincide with higher emission intensity. Labels (a)-(c) correspond to labels in Fig. 2.

Typical regimes of non-equilibrium low-pressure discharges in water vapour (marked with letters in Fig.2 and Fig.3) are:

(a) Low-current Townsend discharge, with exponential increase in emission intensity from the cathode to the anode.

(b) At the beginning of the normal glow, there is a distinct constriction of the discharge, which is followed by the formation of the peak of emission (negative glow), linked to a formation of the cathode fall.

With the increase of the current in normal glow the discharge is broadening radially.

(c) In the abnormal glow, discharge occupies the entire electrode area while the peak of emission is moved closer to the cathode. The increase in discharge current can only be achieved by increase of the voltage.

All of these features are typical for electron dominated processes of excitation and ionization and space charge effects at higher currents. However, at lower pressures, in the left-hand side of the Paschen curve, when reduced electric field (E/N) is high enough, processes of excitation and ionization induced by ions and fast atoms can occur [6]. This is revealed through the additional peak of emission near the cathode (see Fig. 3 for pd=0.3 Torrcm). The most probable candidates for heavy-particles that participate in excitation and ionization are hydrogen ions and fast neutrals [6].

This work has been supported in part by the MPN projects ON171037 and III41011 and bilateral projects Serbia-Slovenia.

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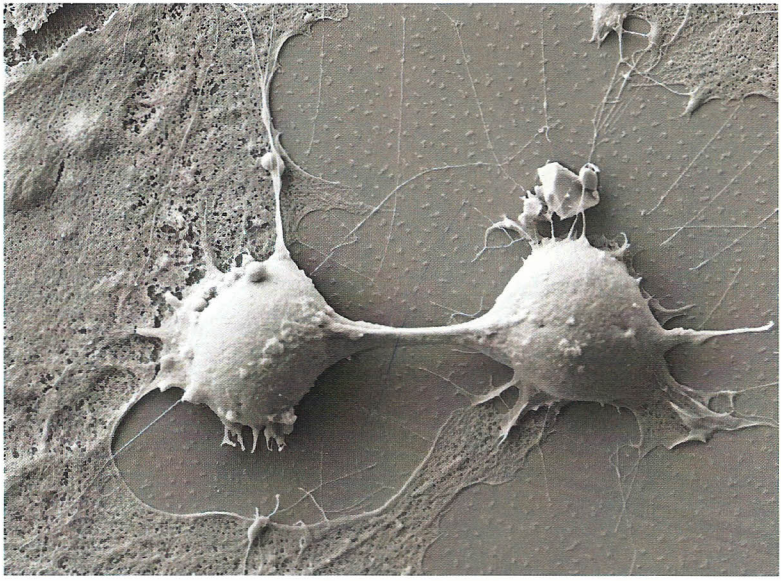
69TH IUVST A WORKSHOP ON OXIDATION OF ORGANIC MATERIALS BY EXCITED RADICALS CREATED IN NON-EQUILIBRIUM GASEOUS PLASMA



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69TH IUVSTA WORKSHOP ON OXIDATION OF ORGANIC MATERIALS BY EXCITED RADICALS CREATED IN NON-EQUILIBRIUM GASEOUS PLASMA

ABSTRACTS

December 9th — December 13th 2011, Crklje na Gorenjeskem, Slovenia

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Electrical breakdown in water vapor and ethanol

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Liquid vapours occur frequently in gas discharges. Vapour is invariably present when plasma is in contact with the living tissue and also when atmospheric gas mixture is used as a component of the gas mixture. Discharges in liquids have become an increasingly active front where often it is claimed that first gas phase is formed before an avalanche may occur or breakdown takes place in bubbles. Finally it is interesting to note that modeling of radiation therapy by electrons or positrons may use the same numerical techniques as used in collisional plasmas [1]. In addition it has been shown that above 5 eV there is little distinction between gas and liquid as due to high speed the wavelength of projectiles becomes smaller than the mean gap between molecules. The cloud of secondary electrons formed when high energy particles are in the gas or in liquid resembles electron avalanche and weakly ionized plasma. Finally, discharges in liquids or gas mixtures containing vapours are often used as detectors of elementary high energy particles.

All of these applications of discharges in liquid may be associated with vacuum technologies albeit very weakly. The strongest point of overlap is that discharges in liquids often lead to production of numerous nano- structures including nano-walls [2].

The breakdown is defining both the pertinent physical processes and operating conditions of plasma. Essential difference between electronegative and electropositive gases arises due to different loss mechanisms, that is projected in the breakdown voltage.

Our apparatus consists of two parallel plates at a gap that may be varied but is typically 1.1 cm. Surfaces are polished copper and we may also use transparent conducting film as an electrode to have a visual recording of the radial profile of

the discharge. We use pulsing of a discharge from the lowest stable current in the Townsend regime to make recordings as a function of the current while we use low current regime and extrapolation to zero current to establish the breakdown voltage [3,4].

The results for water vapour [5] reveal that the breakdown voltage is not strongly affected by the resolved oxygen and purity of water vapour unlike other transport coefficients [6]. It was also observed that an inflection point exists at higher *pd* values (pressure times gap distance) [5]. That point has been observed only for some values of *d* but it occurs at low enough pressures where some phase transitions may be unlikely due to operations close to the dew point. We have observed evidence of a streamer breakdown at some higher *pd* values.

The Volt-Ampere (*VA*) characteristics of the discharge has also been recorded and associated with the operation in different regimes [7]. The spatial emission profiles reveal excitation by H atoms at low *pd* and standard excitation in the region of the minimum and higher *pd* [8].

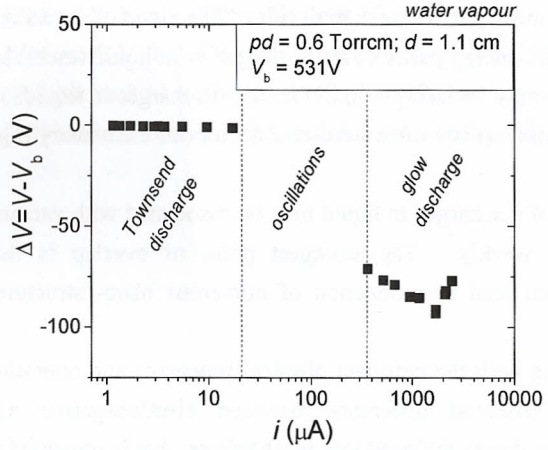

Fig 1 *VA* characterstics of water vapour at *pd* = 0.6 Torr cm [7].

We have made measurements of the Paschen curve in ethanol vapour. The vapour pressure for ethanol at the room temperature is around 40 Torr. We did not observe any inflection points and any irregularities due to dissolved gasses.

The data reveal a very stable Townsend operation up to 0.7 Torr cm and above that point we had to analyze oscillatory patterns to reveal the breakdown voltage. Nevertheless there is no evidence of the transition to the streamer breakdown in this region of *pd* values.

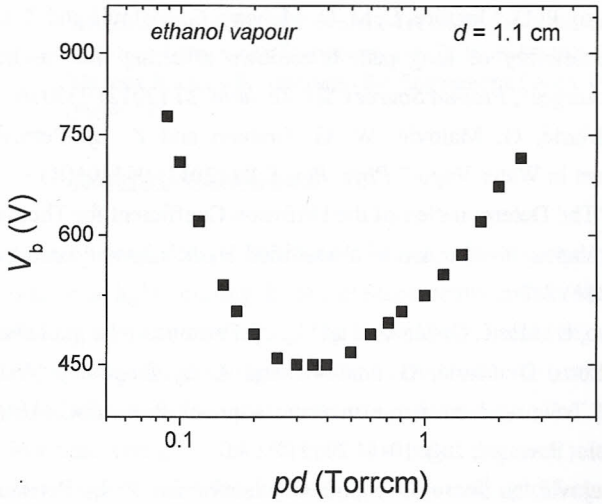

Fig 2 Paschen curve for the breakdown in ethanol vapour [9].

The breakdown in ethanol bears resemblance to that of water vapour as far as spatial profiles go and also for *VA* characteristics. The detailed analysis will be completed and also analysis of the kinetics of processes leading to breakdown will be analyzed.

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Axial emission profiles of Townsend discharge in water vapour

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In this paper we report studies of the basic properties of breakdown and low current Townsend discharge in water vapour in the range of *pd* (pressure x electrode gaps) from 0.1 to 10 Torrcm. Experimental results for Paschen curves at several electrode gaps (from 0.5 cm to 3 cm) and corresponding distributions of emission intensity are analyzed based on elementary processes that are relevant for breakdown and maintenance of low-pressure non-equilibrium discharges.

Increasing interest in application of plasmas in medicine, nanotechnologies and environmental remediation [1,2] has drawn attention to studies of discharges in water and in contact with water [3]. Current studies show that in such systems, discharge is formed in water vapour either from evaporating liquid electrode or in bubbles created by induced phase transition within the liquid. More generally, all atmospheric discharges contain some degree of water vapour. Our aim was to study basic parameters of breakdown in water vapour, which are scarce in literature, and this paper is an extension of our previously published results on Paschen curves for the same gas [4].

In our experiment, the discharge is established in parallel-plate system of electrodes, placed in a tightly fitting quartz tube. The diameter of the electrodes is 5.4 cm. The distance between electrodes is adjustable. In this study, measurements are performed for electrode separations from 0.5 to 3.1 cm. Cathode is made of copper, while anode is a transparent film of platinum deposited on a quartz window. Such construction of the discharge chamber allows us recording of axial and radial profiles of emission. Vapour is introduced into the vacuum system at a slow flow rate. Following the initial period of boiling, water in the tube becomes still and devoid of dissolved oxygen and other volatile constituents. Saturation of all surfaces in the chamber is achieved by containing the vapour in the system for several hours before measurements. Pressure is kept below 20 Torr in order to avoid creation of droplets of water in the discharge chamber.

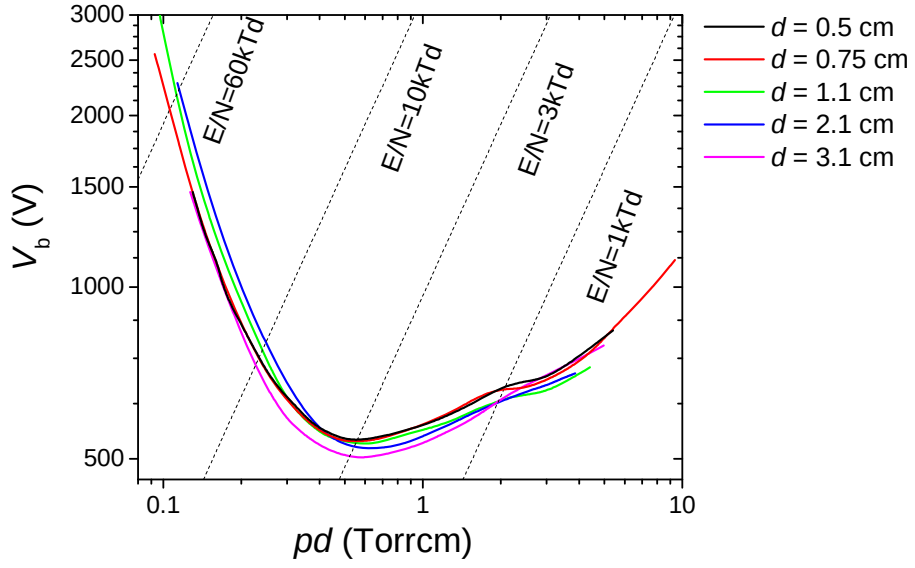

Figure 1 Paschen curves measured for different electrode gaps.

Paschen curve for water vapour is presented in Fig. 1. Breakdown voltage is shown in the *pd* range from 0.1-10 Torrcm for five different electrode gaps. *pd* is standard scaling parameter, proportional to the number of collisions of a particle over distance. Testing of the validity of scaling gives us an insight in processes that participate in breakdown. Reduced electric field (*E/N*) is indicated in Paschen

curve at several points, as another important scaling parameter, which is proportional to mean energy gained between collisions.

In order to obtain breakdown voltages in our experiment, we ignite the discharge in the low-current regime and than by continuously changing the operating voltage we reach the lowest stable, recordable current. Breakdown voltage is then determined by extrapolation of the discharge voltage to zero current. In addition, at the lowest possible current, axial profile of emission is recorded by ICCD camera, to confirm that the discharge is operating in Townsend regime of discharge. Several emission profiles are presented in Fig. 2. We selected profiles at 0.65 Torrcm (Fig. 2a) and 0.11 Torrcm (Fig. 2b) to illustrate how structure of the discharge changes depending on working conditions and reveals basic processes in discharge. As additional test of the validity of scaling, profiles are scaled by *px* (*x*- distance from the cathode), so we may compare emission intensity distribution for different electrode gaps.

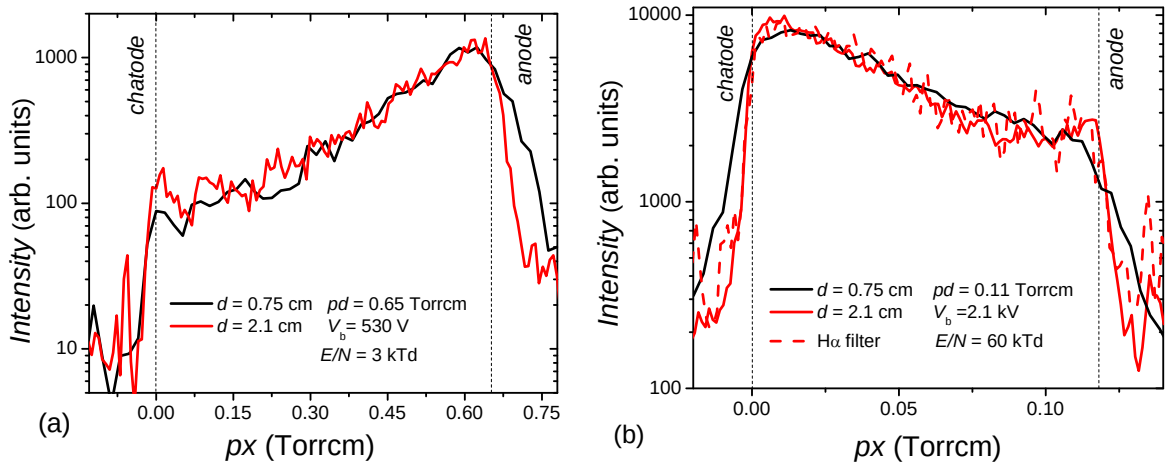

Figure 2 Axial profiles of emission from Townsend discharge in water. Distance from the cathode *x* is multiplied by *p* to enable comparisons of profiles recorded at the same *pd* for different electrode gaps.

In the region of the minimum of the Paschen curve (*E/N* = 3 kTd), we observe typical exponential increase of emission intensity from the cathode to the anode (Fig. 2a). This indicates excitation mainly induced by electrons. Slope of the profile in semi-log scale determines the ionization coefficient of water vapour. From ionization coefficients and Paschen curve we may calculate the secondary electron yield. Thus, measurements presented here give us a set of the key parameters that determine the breakdown [5]. On the other hand, at low *pd*-s (high *E/N*), heavy ions and atoms gain enough energy to perform excitation and ionization and even dominate over electron excitation, which is revealed through emission close to the cathode (Fig. 2b). Fast hydrogen atoms are the most probable candidates for high emission in cathode region [6]. Excellent agreement with the profiles recorded by using H_α filter supports this assumption.

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MONTE CARLO SIMULATION OF ELECTRON TRANSPORT IN H_2O VAPOUR

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Abstract. In this work we present electron transport coefficients in water wapor for the conditions used in plasma assisted technologies. By using Monte Carlo technique appropriate for steady state Townsend discharges we determined mean energy, drift velocity and effective ionization coefficient for a wide range of reduced electric fields (*E/N*= 100-10000 Td). We show that agreement with experimental data for drift velocity and effective ionization coefficients exists for moderate *E/N* implying that one may properly model the electron multiplication.

1. INTRODUCTION

Increasing interest in application of plasmas in medicine, nanotechnologies and environmental remediation [1-5] has drawn attention to studies of discharges in water and with water electrodes [6]. Current studies show that in such systems, discharge is formed in water vapour either from evaporating liquid electrode or in bubbles created by an induced phase transition within the liquid. More generally, all atmospheric discharges contain some degree of water vapour.
It is therefore of immediate interest to determine how discharges are created in water [7–9]. Currently it is thought that discharges can only be formed in water vapor resulting from an induced phase transition. Therefore a starting point must be to have accurate knowledge of the electrical properties of water vapor and in particular its breakdown potential [10,11].

In this paper we study the kinetics of electrons in *E/N* using Monte Carlo simulations (MCS) that have been well tested for similar discharges in Ar, H_2 and N_2. As electrons have a special role in plasmas at high *E/N* we focus on electrons leaving out heavy particle collisions which will be dealt with separately. Another motivation is to provide the transport data for the electrons in the mixtures with H_2O.

2. THE MONTE CARLO TECHNIQUE

The MCS code used in our analysis is based on the null collision method, applied to model a steady state Townsend (SST) discharge. The basic code follows the motion of electrons from the cathode and it is possible to get the spaatial distribution of the emission and the excitation and ionization coefficients [12]. The second MCS code is used to follow the electrons reflected from the anode surface and the newly created secondary electronsfrom the cathode. This code is completely independent from the code for simulation of the electrons from the cathode, and it can be included but also excluded from the simulation. Sampling of observable properties specific to idealized SST experiment between two parallel electrode [13] is applied in order to obtain the mean energy, drift velocity and ionization coefficients. The nonintrusive photon flux experiment of Fletcher [14] and Malović *et al.* [15] provides means of directly observing the periodic structures of electron transport properties which can be classified as representaion of the non-locality of the EEDF and related electron properties in regions where hydrodyic approximation is not fulfilled. In this paper, however, we will present only the data obtained for distances longer than the spatial relaxation length where possible periodic structures vanish.

We used the cross sections data defined in the data base of Phelps [16] initialy based on drift velocity measurements of Pack *et al.* [17]. We deduced elastic momentum transfer cross section from the effective momentum transfer cross section and inelastic cross sections recommended in [16] and extrapolated above 80 eV by using data of Hayashi [18]. Latest recomendation by Phelps [16] for cross sections for ionization measured by Stebbings and coworkers [19] is also applied.

3. DISCUSSION AND RESULTS

In Figure 1 a) we show SST mean electron energy and in Fig. 1 b) SST drift velocity. Latest measurements of drift velocities (mean arrival time

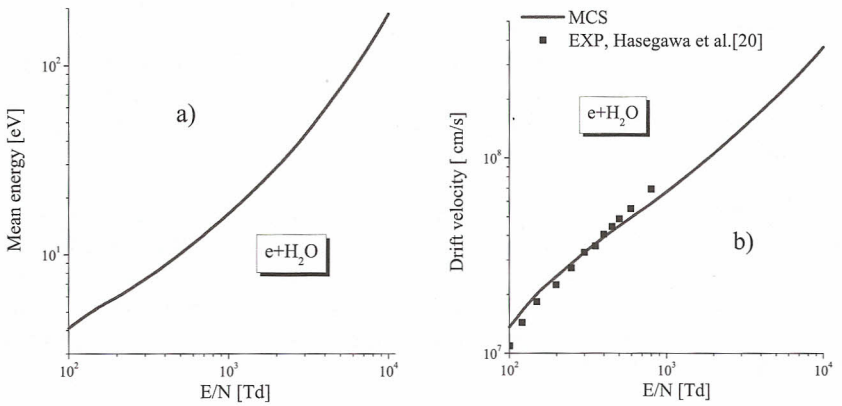

Figure 1. SST a) mean energy, b) drift velocity, obtained by MCS as a function of *E/N*.

drift velocity measurements [9] of Hasegawa *et al.* [20]) are in good agreement with our MCS results.

Figure 2 shows effective Townsend ionization coefficient in the H_2O vapour as a function of *E/N*. Recent ionization coefficients measurements are presented in works of Hasegawa *et al.* [20] recording arrival time electron spectrum and Škoro *et al.* [21] recording side-on spatial profile that show effective charged particle multiplication and is thus under influence of attachiment that reduces the effective ionization. Excellent agreement with experimental data of Hasegawa *et al.* [20] is achived from 150 Td up to 3000 Td while experimental results of Škoro *et al.* [21] are slightly below following the same trend. Beetween 100 and 200 Td the *E/N* dependence of the data due to Prasad and Craggs [22] is followed. Data of Hasegawa *et al.* [20] indicating a possible strong electron loss towards lower *E/N* are not supported by the present cross section set.

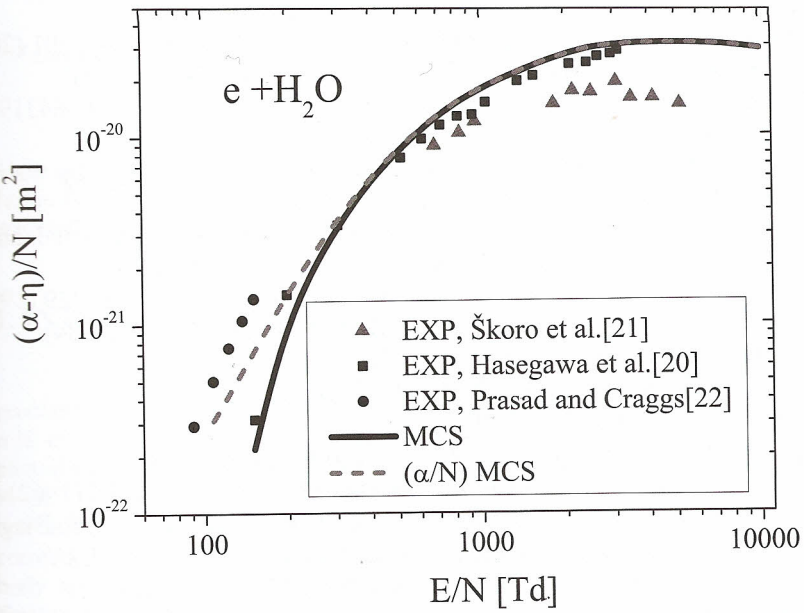

Figure 2. Effective ionization coefficient as a function of *E/N* in water vapor.

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VOLT-AMPERE CHARAC TERISTICS OF LOW-PRESSURE DC DISCHARGES IN WATER VAPOR

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Abstract. In this paper we present the Volt-Ampere (*V-I*) characteristics of a DC discharge in water vapor at 0.6 Torrcm (at the minimum of the Paschen curve) and corresponding emission profiles. Measurements were performed for different electrode gaps (from 0.5 to 3.1 cm). *V-I* characteristics are compared by using standard scaling parameter j/p^2 (j - current density; p- pressure). Some of the issues in proper determination of parameter are discussed. For the gap d = 0.5 cm, a case of coexistence of two discharge channels is presented.

1. INTRODUCTION

Today, many investigations are focused on discharges in liquids (water or some electrolyte), discharges in heterogeneous water-air bubble systems [1] and to atmospheric discharges which operate in ambient air. Many possible applications for treatment of materials, biomedical applications of plasmas [2] and mercury-free sources of lighting [3] are reason for such interest. These discharges operate either in a gas mixture that contains a significant percentage of water vapor or inside vapor bubbles in liquids. It is thus important to know fundamental processes and parameters for pure water vapor discharge ignited in simple geometry. Our goal is to investigate non-equilibrium parallel-plate DC discharge in water vapor with different parameters and obtain a reference set of data (ionization coefficients, secondary electron yield, breakdown voltages) that can be used in modeling of gaseous dielectrics, breakdown, gas discharges and collisional plasmas.

Measurements of breakdown voltages and spatial profiles of DC discharges in water vapor were presented in our earlier paper [4]. Here we present measurements of *V-I* characteristics, which are necessary for a complete analysis of secondary electron production in addition to breakdown data [5].

2. EXPERIMENTAL SET-UP

Discharge chamber consists of two parallel-plate electrodes 5.4 cm in diameter, placed inside a tightly fitting quartz tube. Electrodes separation is adjustable. The cathode is made of copper and the anode is a transparent with a conductive film of platinum deposited on a quartz window. Chamber construction allows us recording of axial and radial profiles of emission. Profiles are recorded by sensitive ICCD camera (Andor IStar DH720-18U-03). Details of the measurement technique are described in [4].

Water vapor, obtained from bi-distilled de-ionized water, is introduced into the discharge chamber from a container through a pressure regulative valve at a slow flow rate. When initial period of boiling is completed, the water in the container become devoid of dissolved oxygen and volatile constituents. In order to saturate the chamber walls, water vapor is maintained at a moderate pressure in the chamber for periods of 1-2 h [4].

3. RESULTS AND DISCUSSION

In figure 1 we show discharge voltage as a function of the reduced current density (j/p^2), for electrode gaps from 0.5 cm to 3.1 cm, at pd = 0.6 Torrcm (at the minimum of the Paschen curve [4]). Voltage is presented as the difference between discharge voltage and breakdown voltage ($\Delta V = V - V_b$), as the breakdown voltage may vary from day to day measurements. At low currents (Townsend regime), the agreement between V-I characteristics at different gaps is good. In normal and abnormal glow discharge, voltages are higher at larger gaps. This is consistent with increased diffusion loses and edge effects at larger gap/diameter ratios and lower pressures [6].

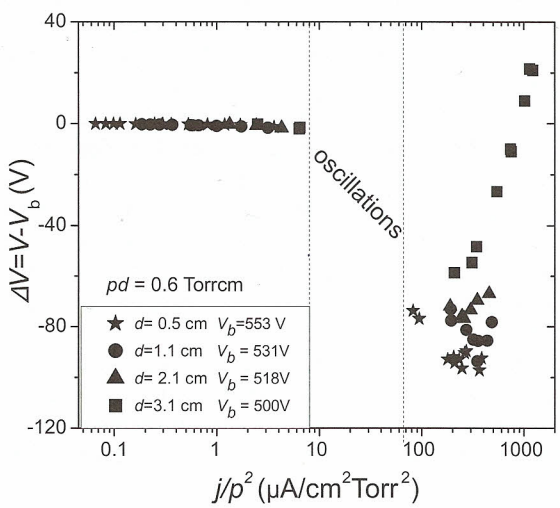

Figure 1. V-I characteristics at pd=0.6 Torrcm for four electrode gaps. Voltage is shown as a difference between discharge (V) and breakdown (V_b) voltage.

One has to be aware of some important issues in determination of the scaling parameter j/p^2. First, when calculating current density from discharge current it is essential to use effective discharge diameter instead of the electrode diameter [7]. We usually determine effective discharge diameter from end-on images of the discharge, recorded by ICCD camera through the transparent anode. However, in measurements presented here, at lower pressures we were not able to record radial (end-on) profiles due to low emission intensities under those conditions and due to limited transparency of the anode. Therefore, we extracted radial profiles of emission from side-on recordings by separating the intensity along the radius of the discharge chamber at the position of the peak of emission. Since in cylindrical geometry, each pixel of the image corresponds to a different volume, depending on the distance from the chamber axis, it was necessary to perform Abel inversion.

Another issue that can be important in determination of j/p^2 parameter comes from the fact that, even at the same *pd,* discharge tends to be more constricted at higher pressures [8]. Constriction is rarely formed at the center of the discharge volume [9]; at certain conditions it can move across the surface [10]; we have even observed the coexistence of double constricted discharge channels. In such cases, estimation of the effective diameter of the discharge is very difficult. There are many possible causes for this kind of complex behavior – if electrodes are not ideally parallel, or if the cathode surface is not homogeneous, if there are high local fields due to roughness of the surface or around the edges etc. The constricted discharge channel will always adjust its position to reach optimal, the most efficient, discharge conditions.

Fig. 2 shows double channels of the discharge that were observed in Townsend and normal glow regime. Axial profiles of emission (at d=0.5 cm) along the channels and corresponding 2D images of discharge are presented. The emission intensity of the secondary channel is lower, but similarity of the profiles proves that channels operate in the same regime. In order to make comparison of profiles, maximum intensities of secondary channels are normalized to the maximum intensity of corresponding main channels. It is interesting to note that profiles for different currents are normalized by the same factor, which means that relative intensities in dual channels are preserved in different regimes of the discharge. The coexistence of two channels in the discharge could be related to a small diffusion length at higher pressures and large electrode diameter (compared to the gap), where constrictions can operate independently [8,9].

In the case presented here, effective discharge diameter can be estimated as a sum of diameters of the two channels. However, it is difficult to claim that such estimation is realistic, as distribution of the electric field along the channels may not be the same.

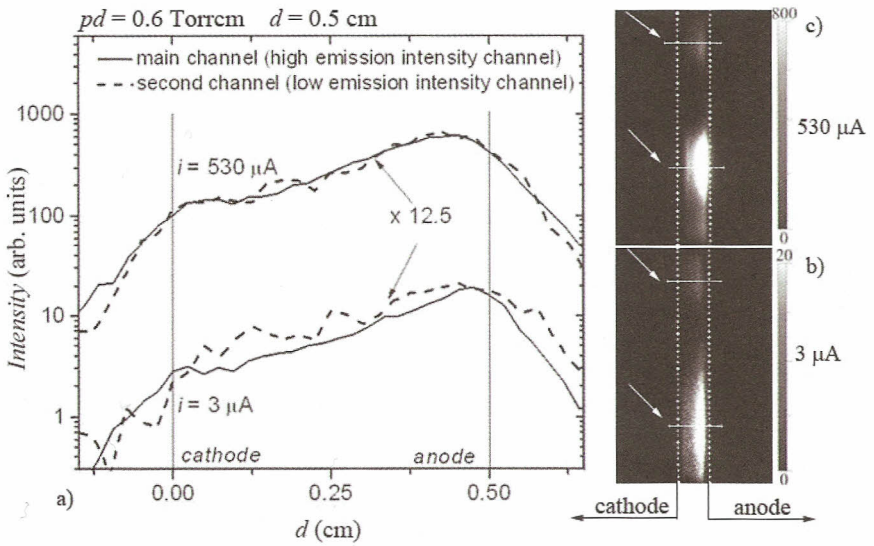

Figure 2. Profiles of emission along the constricted channels (a), indicated by solid white lines at corresponding 2D images (b, c) in Townsend regime of the discharge (3 μA) and in normal glow (530 μA).

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Contents

Invited Lectures

BREAKDOWN AND DISCHARGE DEVELOPMENT IN VARIOUS GASES AND ELECTRODE CONFIGURATIONS

Dragana Marić[1], Jelena Sivoš[1], Nikola Škoro[1], Gordana Malović[1], Thomas Kuschel[2], Ilija Stefanović[2], Joerg Winter[2] and Zoran Lj. Petrović[1]

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In this paper we give an overview of the most important aspects of the influence of various gases, electrode configurations and dimensions to the gas breakdown and the discharge development. Paschen curves in parallel-plate geometry are shown for a number of atomic and molecular gases. Measurements with hollow-cathode configuration enabled us to establish a relation between Volt-Ampere characteristics and space-time resolved development of the discharge structure. Finally we emphasize some of the main issues in application of scaling laws and interpretation of results in micrometer-size discharges.

1. Introduction

After more than a century, electrical breakdown still remains an attractive topic for studies in the field of physics of ionized gases. With improvements in experimental and modelling techniques, over the past 20 years there has been a significant advance in resolving some of the long standing issues and generally in understanding of processes and phenomenology of breakdown and development of discharges. Phelps and his coworkers have revised standard Townsend's theory of breakdown and low-current discharges [1-4]. Revision included all possible feedback mechanisms and effects of space charge, so it was possible to explain negative differential resistivity and oscillations in low-current Townsend's discharge. It even led to explanation of oscillations in the low current diffuse regime [1,2] and of constrictions of the discharge in the higher-current normal glow discharge [3,5]. Furthermore, procedure that was recommended for determination of secondary electron yields in low-current discharges [4] has produced data that could be used up to the abnormal glow discharge [6].

Motivation for this work comes from the need to achieve a good control of the breakdown and the discharge development and to gain the knowledge of the elementary processes and their kinetics. Ionization in the gas phase and secondary emission/surface interactions are in focus of our studies as the main mechanisms of breakdown and discharge sustainment. In addition we believe that implementation of elementary processes needs to be tested from both the low current end and also at higher currents. As one may often assign different processes under one simplified agent, for example flux of ions is used to represent all mechanisms of production of secondary electrons at the cathode, one needs to test how such assumptions would fare at regimes at higher currents where linearity between different fluxes and the electron density may be broken. Laboratory for gaseous electronics at the Institute of Physics Belgrade has a long experience in research of low-pressure dc non-equilibrium discharges realized in parallel-plate geometry. This enables us to extend our knowledge and experience to more complex geometries, to small dimensions and high pressures, and to high frequency discharges. Our usual approach is to test assumed processes and test possibilities of the entrance of new physics. Finally, some of the plasma applications depend directly on breakdown, while others require referent data sets for further development and optimization.

In this paper, we will start from the results obtained for various gases in a simple parallel-plate geometry at standard pressures and dimensions (p~1 Torr, d~1 cm). Further on, we analyze discharges in hollow-cathode geometry. Finally, we will show how reduction of discharge dimensions, with an increase in pressure, influences properties and behaviour of the discharge.

2. Breakdown in various gases

Breakdown is usually represented by the Paschen curve i.e. dependence of the breakdown voltage on the *pd* (pressure *p* x gap *d*). *pd* is a scaling parameter proportional to the number of collisions over a certain distance. In this respect, a typical sharp increase of the breakdown voltage at low *pd*-s can be explained by the need to compensate for a small number of collisions. On the other hand, at high *pd*-s, due to a large number of collisions, breakdown voltage is increased in order to enhance the energy gain between collisions, when mean free path is getting shorter. In the range of the Paschen minimum, production of charges by ionization and secondary electron emission and losses by diffusion and drift are well balanced.

There are several issues that one has to be aware of in breakdown measurements. Breakdown voltage depends on the gas or the mixture and on the cathode material. But even more important than the cathode material is the state of the cathode surface – possible oxide layers and other impurities deposited on its surface either by exposing the cathode to the laboratory environment or during the discharge operation. At micro dimensions roughness of the surface will play and increased role. Sometimes the state of the cathode surface has a greater influence on Paschen curve than the material of the cathode itself. For this reason, in our experiments, cathode surfaces are treated in low current (~30 μA) hydrogen discharge prior to the breakdown measurements [6]. This procedure proved to give stable conditions during measurements and reproducible results over long periods of time.

Another issue that has to be taken into account in experiments is the regime in which the discharge ignites. Breakdown voltage should not be confused with the operating voltage. The point where the discharge ignites is at the crossing of the circuit load-line and the Volt-Ampere characteristics. Following the breakdown the discharge makes often a very rapid transition to the operating voltage best suited to the Volt-Ampere characteristics and the external circuit (load line). Quite often, especially with small circuit resistance and large overvoltage, this is in the range of glow discharge, where voltage can be significantly smaller than the breakdown voltage. Actual breakdown voltage, in the sense that is represented by the Paschen law, can be found by extrapolating Volt-Ampere characteristics to zero current in dark Townsend discharge mode, but not from the glow discharge. While operating voltage plotted against *pd* may have the same dependence as the Paschen curve the physics of the glow discharges and the fact that in different *pd* regions final state may be different indicate that such plots cannot be used to obtain secondary electron yields and to represent the breakdown.

In Figure 1 we show Paschen curves for several atomic and molecular gases. Measurements with N_2, SF_6, CF_4 and water vapour are taken with the copper cathode, with 1 cm electrode gap and 5.4 cm diameter. For all other gases, stainless steel cathode was used in measurements in 1.72 cm gap and 8 cm electrode diameter.

For most of the gases, Paschen minimum is situated at *pd* of the order of 1 Torrcm and breakdown voltages are of the order of several hundred volts. In the case of electronegative gases, position of the minimum is usually shifted towards smaller *pd*-s and higher voltages. This can be understandable from the point of view of the balance of production and losses of charged particles. In electronegative gases, at low *E/N* i.e. high *pd*, attachment becomes important. As a loss mechanism for electrons, it will increase the breakdown voltage and shift the Paschen minimum to lower pressures as an even higher *E/N* is required to provide sufficient ionization.

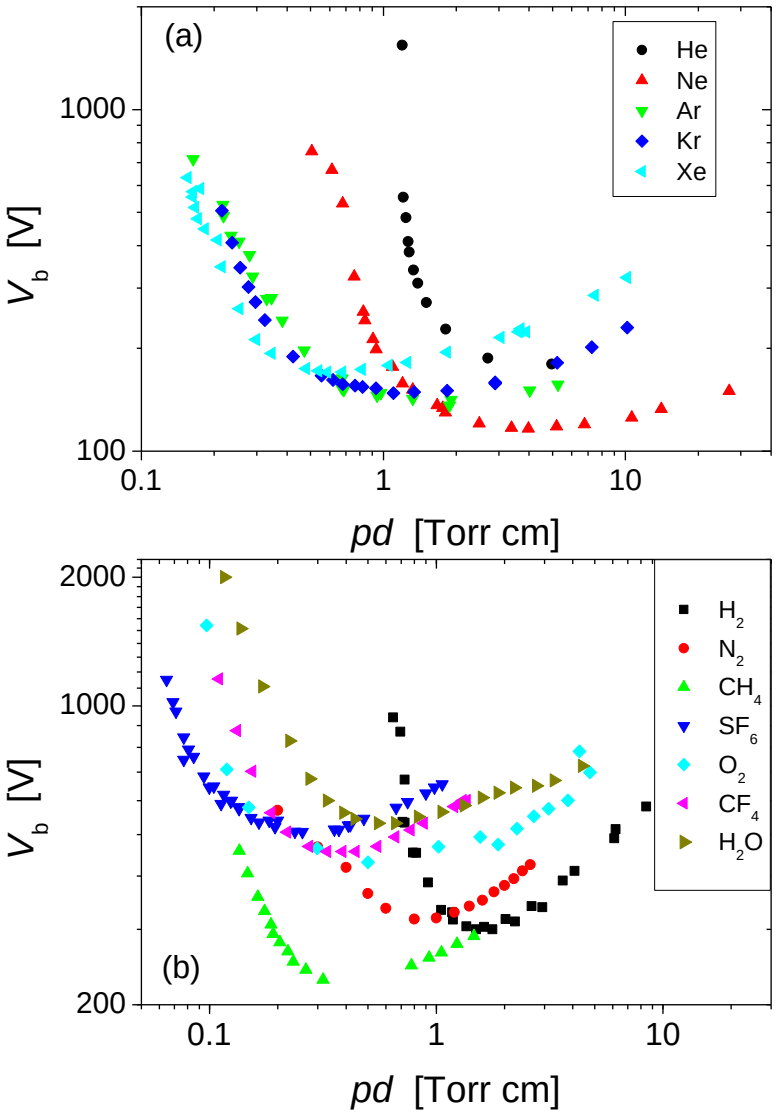

Figure 1 Paschen curves for selected (a) atomic and (b) molecular gases [7,8].

In our experiments, measurements of the breakdown potentials are always supported by recordings of the distribution of the light emission by ICCD camera. That way, we may verify the regime of the discharge from the profiles of emission. In low-current Townsend discharge, close to the breakdown, intensity of emission typically increases exponentially from the cathode towards the anode. In addition, from the slope of the profile we may determine the ionization coefficients. From the data for breakdown voltages and ionization coefficients, we calculate secondary electron yields, based on the breakdown condition. Secondary electron emission coefficients obtained this way are effective - they include all feedback processes and they are valid for exact cathode surface conditions and purity of the gas used in the measurement. [4,8,9]

It is important to emphasize that, besides the Paschen curves, voltage-current characteristics are essential in understanding the process of breakdown. These data are crucial in verifying models of secondary electron emission and gas phase multiplication in order to be applied in comprehensive modelling of higher current plasmas. The slope of the characteristics is negative in low-current region and it reveals the ion energy dependence of the secondary electron yield [1,2,10]. In practice, for a full description of the discharge, we construct 3D plot, such as one shown in Figure 2, with discharge voltage (*V*), pressure x electrode gap product (*pd*) and discharge current (*i*) presented at axes.

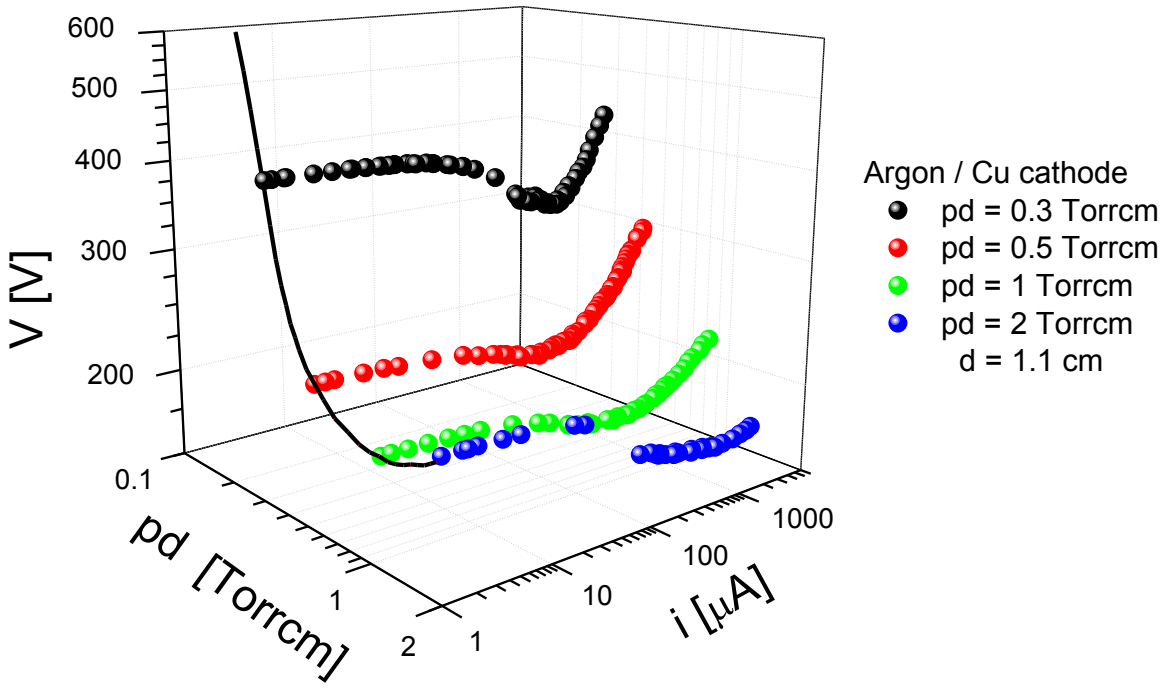

Figure 2. Dependence of the discharge voltage (*V*) on the pressure x distance between electrodes (*pd*) and discharge current (*i*) for argon. Low-current limit represents the Paschen curve (black line) [8].

3. Discharges in hollow-cathode geometry

More than 80 years of studies of hollow-cathode discharges have brought many applications in light sources [11], analytical spectroscopy [12,13], and analysis of nanostructured surfaces [14]. A whole new field of research recently emerged, with miniaturization of hollow-cathode discharges. It brought a number of new possible applications, especially in plasma processing [15]

Advantage of the hollow-cathode over parallel-plate geometry is mainly in efficiency – hollow-cathode discharges have lower breakdown voltages and they operate at lower voltages for the same current density, due to a pendulum motion of energetic electrons and ions [16-18]. The pendulum motion is induced by spatial profile of electric field penetrating the hole in the cathode and thus the electron path is extended, giving more chances for ionization and allowing operation at lower pressures with reasonably small voltages. In addition, in the glow regime of the discharge, when negative glow regions of opposing cathode surfaces overlap, a "hollow-cathode effect" appears. This effect is manifested through sudden increase in current density and intensity of light emission, with decrease of the discharge voltage [17-20].

Although basic processes in hollow-cathode discharges are fairly well known, recent studies of micro hollow-cathode discharges have opened some new issues [21,22]. It was found that for these discharges, the hollow-cathode effect is not significant, due to the short mean free path at higher pressures that are employed in experiments. Furthermore, the results raised the issue of the scaling of discharge properties. The main problems are in proper determination of effective path of the discharge (determination of the *pd* scaling parameter) and determination of effective diameter of the discharge (j/p^2 parameter). It became obvious that it is essential to make a clear relationship between electrical characteristics and spatial structure of the discharge in complex electrode geometries.

We have made measurements of Volt-Ampere (*VA*) characteristics of low-pressure (2 Torr) cylindrical hollow-cathode discharge in neon, supported by ICCD imaging [20,23]. Measurements were done for steady state conditions, but also in transient phases of the discharge formation. Our analysis was based on comparisons with simpler parallel-plate discharges [24]. Figures 3 and 4 show *VA* characteristics and radial images of the discharge for several selected points respectively. Images are recorded through the ring anode.

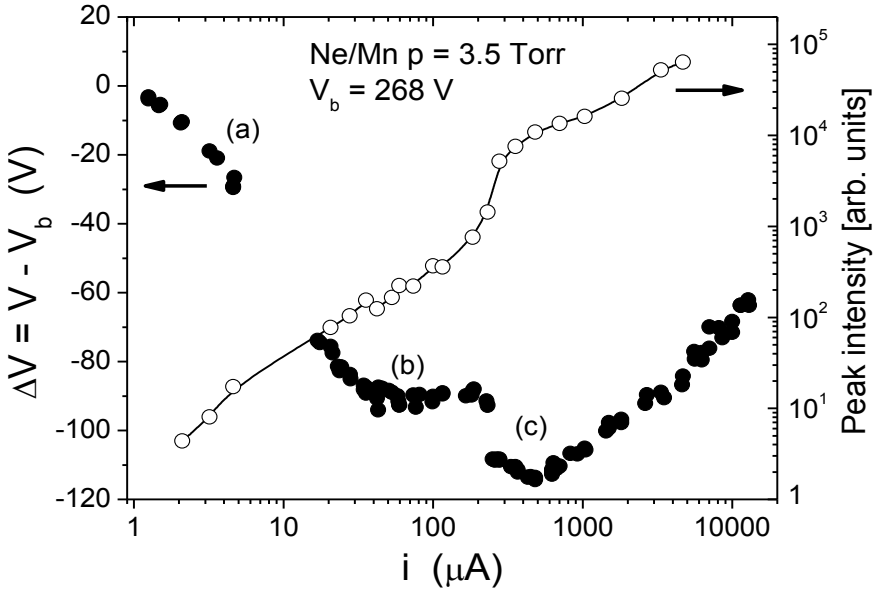

Figure 3. Volt-Ampere characteristics of the hollow-cathode discharge (solid circles) and peak intensity of emission, taken at the center of the discharge (open circles).

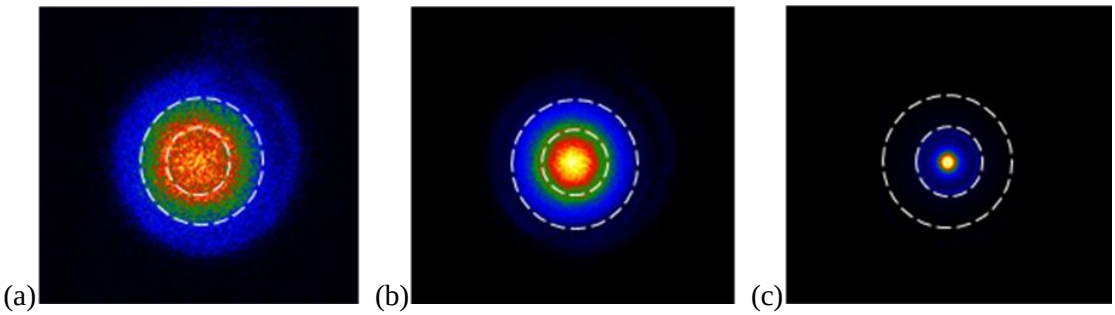

Figure 4. End-on images of the hollow-discharge. Labels (a)-(c) correspond to the labels in the figure 3 [20,23].

Figures 3. and 4. show that in the low-current limit, the discharge resembles the Townsend regime in parallel-plate geometry (label (a)). The *VA* characteristic has typical negative slope and region of oscillations, while the discharge is completely outside of the cathode cavity – it operates between outer surface of the cathode and the anode. Label (b) marks a region of the discharge can be related to the parallel-plate normal glow. Voltage becomes constant with the increase of current. In this range, the discharge operates both, between electrodes and inside the cathode cavity i.e. the voltage remains constant at the expense of extending the discharge area. Normal glow-like regime (b) is followed by a sudden drop of voltage with a large increase of emission intensity at the centre. The discharge now operates entirely inside a cathode cavity. It has switched to the more efficient mode of operation, typical for "hollow-cathode effect". Further increase of current requires increase of voltage, while the discharge remains inside the cavity. Time-resolved measurements have shown that transition to the hollow-cathode mode of operation is not followed by instabilities (oscillations); the discharge

continuously adjusts itself to the more efficient regime over the period of few hundreds of microseconds to several milliseconds [20,23].

Presented results gave us the insight in influence of electrodes geometry to the properties of the discharge. Complementing measurements with model such as one described in [19] would certainly improve the analysis of elementary processes that determine behaviour of the discharge. It is important to note that a proper analysis of the scaling laws in hollow-cathode discharge requires extending the measurements with recordings of axial distributions of the emission intensity. Without those measurements, it is difficult to determine scaling parameters properly.

4. Breakdown of micro-discharges

As noted in the introduction of the paper, micro-discharges have attracted a lot of attention in physics of ionized gases lately. This is mainly due to a possibility to achieve non-equilibrium conditions even at atmospheric pressure, which opened a road to development of many new applications [15,25-27]. Miniaturization of the discharge gives us a possibility to operate at the conditions near the Paschen minimum due to *pd* scaling law. Furthermore, the best approach to studies of micro-discharges is to start from the low-pressure discharges and employ scaling laws [6,28].

Modelling results have shown that *pd* scaling should be valid down to the electron gaps ~10 μm [29]. At lower gaps field emission becomes the main mechanism of electron production, which leads to the sharp decrease of breakdown potential with the decrease of *pd* in the left-hand branch of the Paschen curve. However, breakdown voltages appear to depart from the standard Paschen curves even at gaps of the order of 100 μm [28]. In figure 5, Paschen curves for several electrode gaps in this range are presented. It was found that lowering of the breakdown voltage at low *pd*-s is due to the long-path breakdown [30]. In a strict sense, this is not a violation of the scaling law, as the decrease of the voltage is a result of the increase of effective discharge path. However, results imply that measurement of the left-hand side of the Paschen curve is not possible in open structures [28,30].

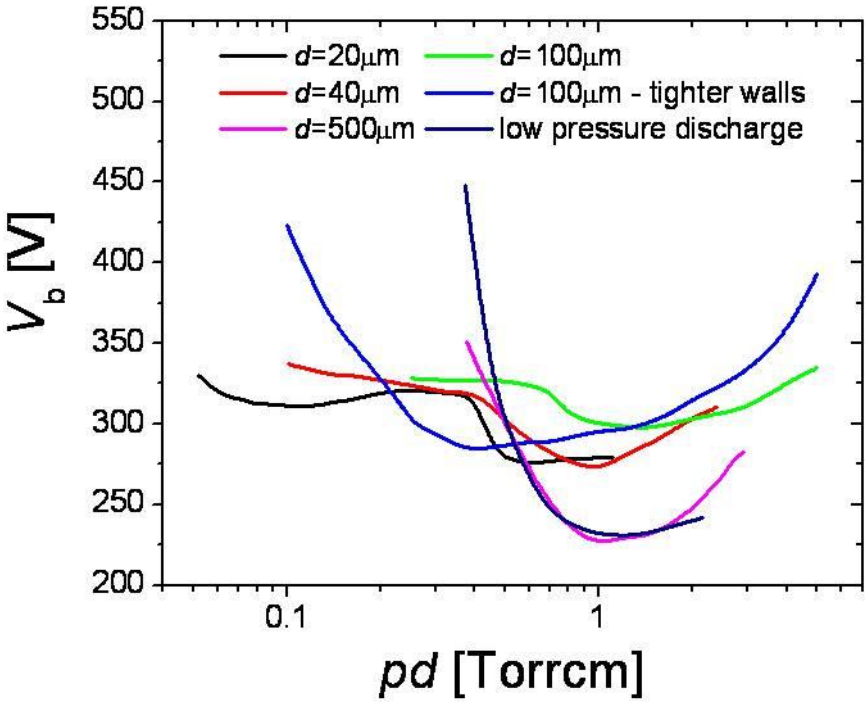

Figure 5. Paschen curves for a parallel-plate dc micro-discharge at several electrode gaps, compared to the low-pressure centimetre-size discharge [28].

Effects of the long-path breakdown can be related to the breakdown in complex geometry of electrodes, where electrode gap is not the same in entire discharge volume. We have measured the Paschen curve in micro hollow-cathode geometry (Figure 6) [30]. Cathode was closed-end cylinder, with anode plate placed at 500 μm distance. Glass plate coated with transparent ITO film was used as the anode, so it was possible to record radial structure of the discharge by ICCD camera. Selected 2D radial distributions of emission intensity are presented in figure 7. Figures 6 and 7 show that at lower *pd*-s the discharge is established between the anode and the inner surface of the cathode cavity (a) i.e. discharge chooses the longest possible path, that will effectively reduce the breakdown voltage. As the *pd* increases, the discharge gradually changes its path and at higher pressures it operates along the shortest part of the gap (c). It should be noted that in figure 6 the breakdown voltage is normalised to the value that corresponds to the Paschen minimum in parallel-plate case. This was done to make a better illustration of the effect, but overall, voltages are shifted to higher values in complex geometry due to additional diffusion losses of electrons and ions to the parts of the volume that are not occupied by the discharge [30]. Similar behaviour can be observed in figure 5, under the conditions where long-path breakdown occurs.

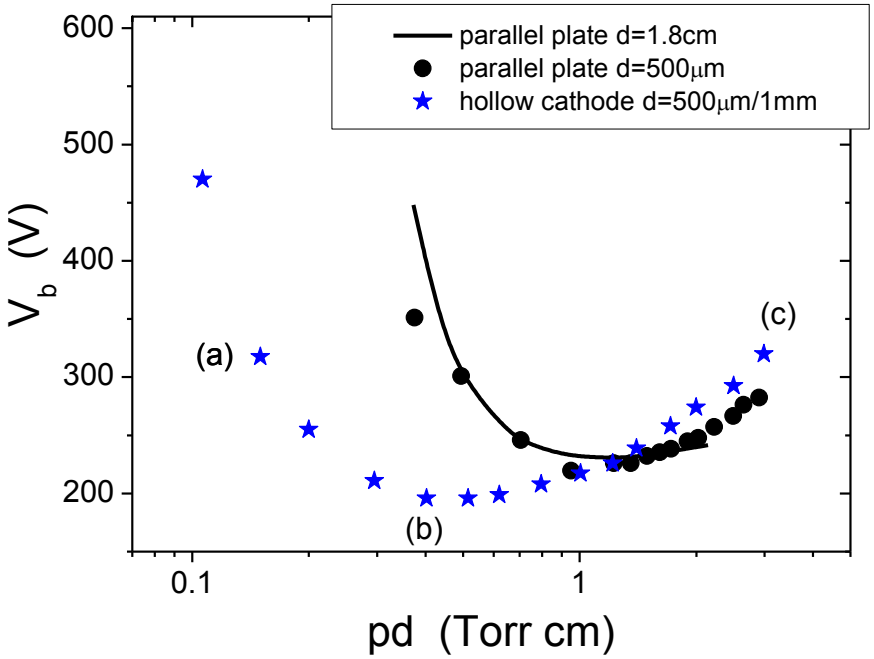

Figure 6. Paschen curve for micro hollow-cathode discharge, compared to the parallel-plate breakdown curve [30].

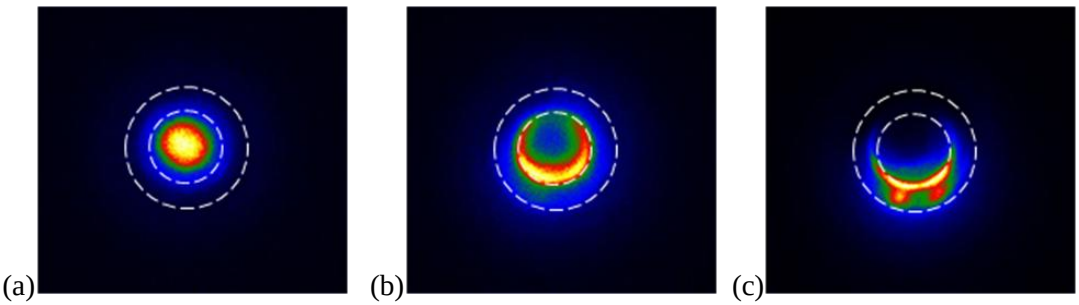

Figure 7. 2D end-on images of the micro hollow-cathode discharge. Labels (a)-(c) correspond to the conditions indicated in figure 6 [30].

Small changes in gas flow also have significant influence on Paschen curve. Figure 8 presents Paschen curves for parallel-plate micro-discharge measured with gas flows from 5 to 50 sccm. It seems straightforward to assume that observed differences are a consequence of (*i*) the pressure gradient inside the discharge volume or (*ii*) removal of charged particles from by the influence of flow. However, in order to test the first assumption, pressure was measured at two points – right before the

entrance of the gas to the discharge chamber and at the exit point, towards the vacuum pump. These measurements confirmed that the differences in results cannot be contributed to pressure gradients. In order to test assumption (*ii*), velocity of gas through the discharge chamber was estimated from the gas flow and the electrode gap, as a narrowest section through which the gas flows. Estimated velocity is several orders of magnitude lower than drift velocities of electrons and argon ions [4]. Therefore, it seems improbable that the removal of charged particles will have significant effect. Examination of other possible factors that could explain presented results is in progress.

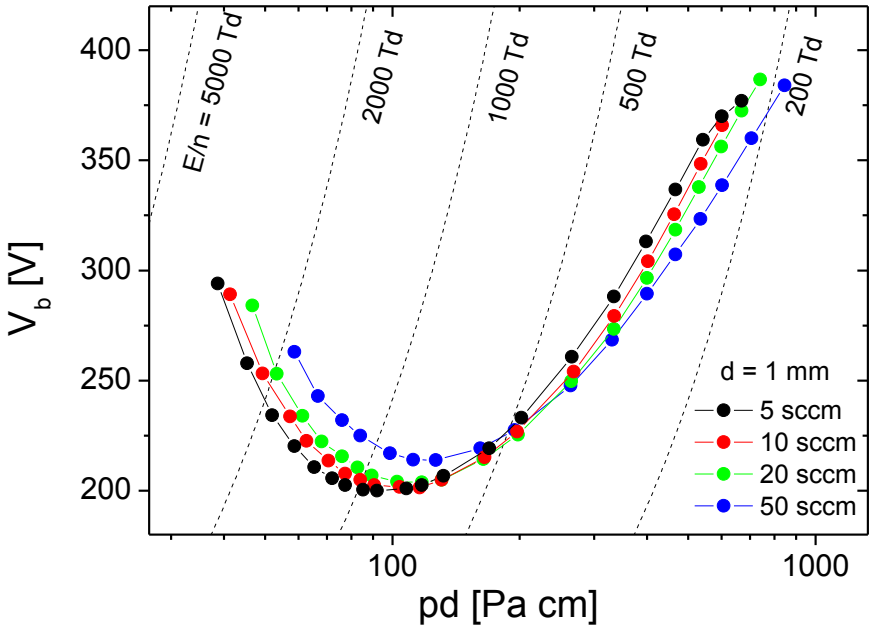

Figure 8. Paschen curves for different gas flows of argon [31].

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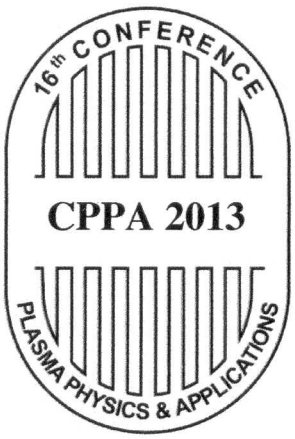

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Electrical breakdown in low-pressure ethanol vapour

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Discharges in liquids and in vapours of liquids are an increasingly active field of research. Vapours are invariably present in plasmas for living tissue treatment and generally in plasmas that operate in atmospheric gas mixtures. Recently, discharges in organic liquids proved to be an efficient source for high-speed production of graphene layers [1]. Furthermore, mixtures of gases in high energy particle detectors contain organic vapours [2]. In all cases, studies are limited by the lack of relevant data on elementary processes. Breakdown studies can reveal the information on processes and their balance in discharges. We will present the breakdown data for ethanol vapour, in comparison to the recently published results for water vapour [3].

Paschen curves (breakdown voltage vs. pressure x electrode gap) for ethanol vapour and water vapour are presented in figure 1. Details of our measurement technique are described in [3, 4]. Over the entire range of operating conditions, the pressure is kept below characteristic vapour pressures (at room temperature ~40 Torr for ethanol vapour; ~20 Torr for water vapour) to avoid formation of liquid droplets. Also, there is no transition to streamer breakdown for the investigated range of *pd*. Minimum of the Paschen curve for ethanol occurs at 0.35 Torrcm, which is somewhat lower than for the case of water vapour (0.6 Torrcm). At $pd < 1.5$ Torrcm, voltages required for breakdown are lower in ethanol vapour. More detailed analysis of the processes that contribute to the breakdown will be presented, based on recorded spatial emission profiles.

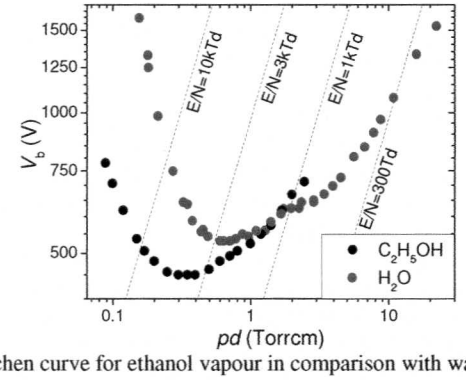

Figure 1. Paschen curve for ethanol vapour in comparison with water vapour [3]

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Breakdown in water vapour and ethanol vapour: heavy particle processes

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Abstract In this paper we report studies on breakdown and low-current discharges in vapours of water and ethanol. Special attention is devoted to processes induced by heavy particles – positive ions and fast atoms and molecules. Contribution of heavy particle processes to the discharge is based on analysis of axial profiles of light emission. Spatial profiles are recorded by ICCD camera, which is sensitive in visual part of spectra. Furthermore, optical filters are used to obtain additional information on dominant processes in breakdown. Obtained results show that, within visual range of spectra, major contribution to the emission comes from atomic hydrogen excitation, while in ethanol vapour this role belongs to heavier particles produced in ethanol dissociation, such as CO, CH and CH_xO.

INTRODUCTION

Non-equilibrium gas discharges play an important role in nowadays technological development. They become an indispensable part of microelectronics manufacturing, material processing, environmental, bio- and nano-technologies. Particularly, water discharges draw attention with wide range of applications in plasma medicine, light source improvement and environmental remediation [1,2]. On the other hand, ethanol is attractive as a potential fuel of the future [3]. Also it is shown that ethanol can be used for growth of carbon nanostructures [4]. As examples of simple liquid compounds, water and ethanol are suitable for investigations of elementary atomic and molecular processes in discharges, which are inevitable step in further development of applications.

EXPERIMENTAL SET-UP

Breakdown and low-current discharges are obtained between two parallel-plate electrodes, placed inside a tightly fitting quartz tube. Copper cathode and quartz anode coated with transparent, conductive platinum film are 5.4 cm in diameter. Electrode gap can be varied between 0.5 and 3 cm. Axial profiles of light emission are recorded using sensitive ICCD camera (Andor IStar DH720 - 18U - 03) with a photo-objective transparent in a visual spectral range. For preliminary spectrally resolved measurements of light emission we have used two optical filters. The narrow pass (Carl Zeiss JENA) Hα filter with maximal transmittance at $\lambda = 655$ nm is used for recording the Balmer emission of H atoms. The band pass Schott BG25 filter, with a transmission greater than 50% between 330 nm and 470 nm, and around 30% at wavelengths above 700 nm is filtering emission lines from dissociation products of ethanol molecules.

Ethanol vapour is obtained from 95% ethanol (C_2H_5OH), while water vapour was produced from bi-distilled and deionised water. The vapour is introduced into the system from a test tube with liquid sample through a pressure regulatory valve at a slow flow rate. This leads to an initial period of boiling water/ethanol, which then becomes devoid of dissolved gases. After few minutes, the surface of liquid becomes still, while vapour pressure becomes stable. Water and ethanol vapours are maintained at a moderate pressure in the chamber for periods of 1–2 h in order to saturate the chamber

walls before measurements. The vapour pressure for ethanol, at room temperature, is around 40 Torr and for water it is 20 Torr. During the measurements, we keep operating pressures below these values to avoid formation of liquid droplets.

After breakdown, the electric circuit with large resistance enables operation of the discharge in stable regime at low current. The breakdown voltage is obtained from the low-current limit of the discharge, in other words, by extrapolating the discharge voltage to zero current in voltage-current characteristic.

RESULTS AND DISCUSSION

In Figs. 1 and 2 axial light emission profiles of discharge in water and ethanol vapour are shown. The profiles correspond to the low-current limit of the discharge, i.e. Townsend discharge (~1 μA). Emission profiles were recorded in full visual spectra and with band-pass filters. Transparency of filters is taken into account, so intensities are relatively scaled. Very high reduced electric fields (*E*/*N*) are attained in the left branch of Paschen curve. Profiles recorded for moderate *E*/*N* are obtained around minimum of the Paschen curve for both vapours. Narrow-pass Hα filter is used to identify emission from excited H atoms. In the case of ethanol discharge, filter Schott BG25 is also used as a preliminary test of emission from several species, which are expected to form through dissociative excitation in ethanol vapour (CO, CH, CH_xO) [6].

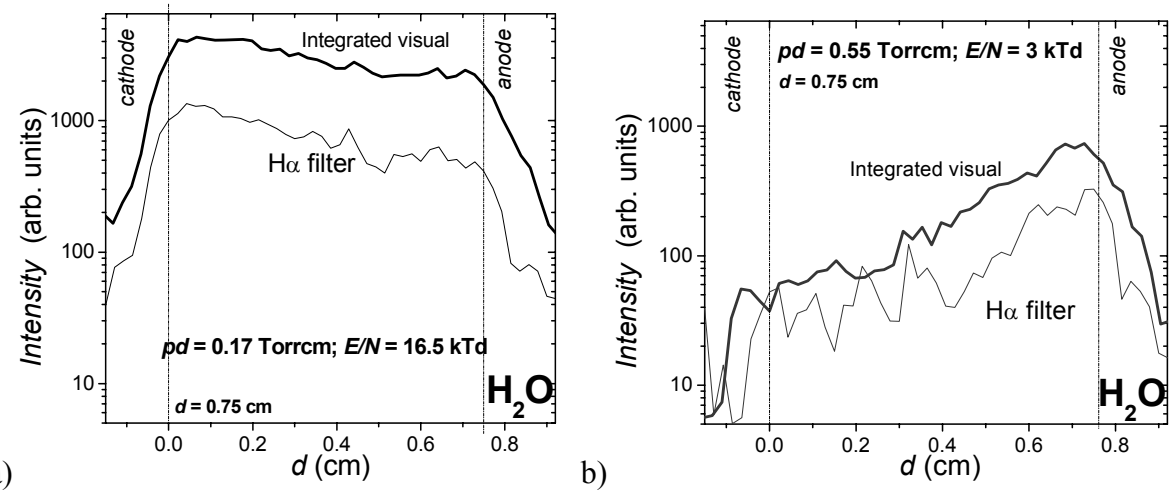

Fig 1. Axial emission profiles of water vapour discharges at 16.5 kTd (a) and 3 kTd (b). Discharge parameters and filter designations are noted in the plots.

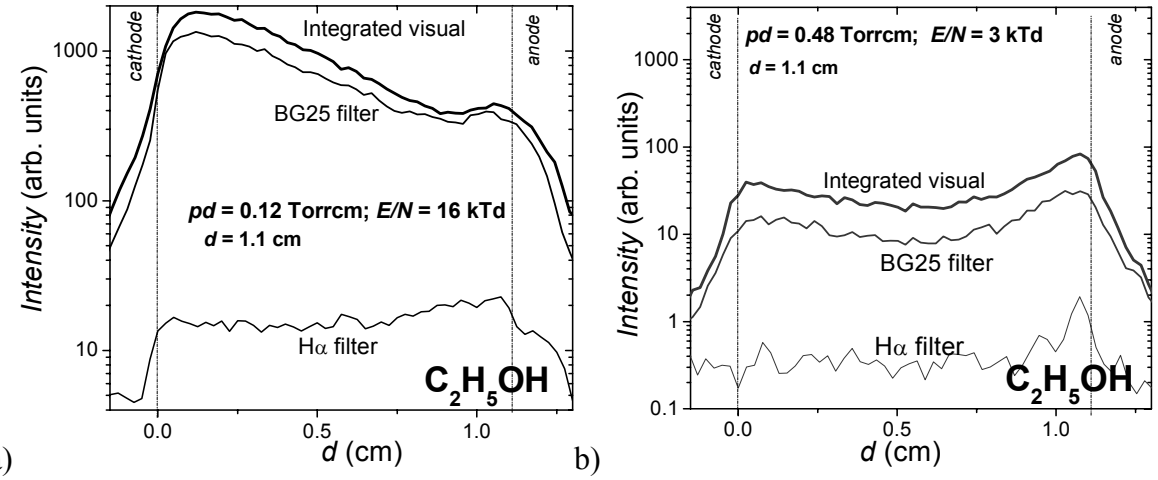

Fig. 2. Axial emission profiles of ethanol vapour discharges at 16 kTd (a) and 3 kTd (b). Discharge parameters and filter designations are noted in the plots.

At high *E/N*, profiles in visual spectra of both vapour discharges are peaking towards cathode as a consequence of significant contribution of heavy-particle processes in the vicinity of the cathode [5]. Filtered emission can reveal ions, atoms and molecules involved in excitation and ionization by electrons and heavier particles.

In water vapour (Fig. 1a), shape of the Hα emission profile follows the integrated visual profile. The filtered profile intensity is only ~3 times smaller than the profile in visual spectra, indicating that a large part of emission comes from excited hydrogen atoms produced in the discharge. In the case of lower reduced electric field (Fig. 1b), Hα profiles again follow the shape of integrated emission intensity distribution in visual part of spectra, but with typical exponential increase of intensity towards the anode due to electron-induced excitation and ionization of particles in the discharge [5, 7]. Difference between profile intensities is small, thus leading contribution to overall emission is by Balmer α line.

In ethanol discharge, at high *E/N* (Fig. 2a), contribution of Hα emission is significantly smaller and the shape of the profile does not follow the overall visual emission. At lower *E/N*, Hα emission is hardly distinguishable from background noise. Emission from heavier dissociative products of ethanol (CH, CO, CH_xO, O) makes dominant contribution. Furthermore, processes of excitation by ions and fast neutrals in the vicinity of the cathode are more pronounced in ethanol than in water vapour. Even at lower *E/N* (Fig. 2b) there is a notable emission in the region close to the cathode. In order to distinguish between different processes that contribute to emission in the range from 400 to 450 nm, further spectrally resolved measurements are planned.

CONCLUSIONS

Emission profiles of low-pressure water and ethanol vapours were recorded in steady-state discharges running at very low currents. The spatial profiles were recorded integrated in visual spectra and with band-pass filters. This allowed us to determine characteristic particles involved in the emission from the discharge volume. At high *E/N*, the electric field is strong enough to accelerate atoms and radicals in the cathode region to allow excitation induced by heavy particles. In case of ethanol vapor discharge, carbon containing species and possibly atomic oxygen were identified as dominant target particles involved in heavy-particle processes. In water vapour, atomic H plays major

contribution. At lower electric fields, electron excitation is dominant process since profiles are peaking close to the anode. Nevertheless, in ethanol discharge strong emission is observed near the cathode even at lower values of reduced electric field.

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P-FUN6
BREAKDOWN IN ETHANOL VAPOUR

J. Sivoš, N. Škoro, D. Marić, G. Malović, Z. Lj. Petrović

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Many studies are currently directed to the discharges in ethanol and ethanol vapour. It is one of the most prospective fuels for the internal-combustion engines [1], as a clean and renewable source of energy. The discharges in ethanol also proved to be an efficient source for high-speed production of graphene layers [2]. In a broader view, vapours of organic liquids are present in mixtures of gases used in detectors of high energy particles; in plasmas for living tissue treatments; in some planetary and satellite atmospheric plasmas. All of those include very complex chemistry, while relevant data are mostly lacking even for the pure constituents of mixtures of gases and vapours. As one of the simplest organic compounds, ethanol vapour is suitable for studies of elementary processes and their kinetics. Those studies can be extended to understanding of more complex systems. One of the aims is to provide reference data for modelling.

In this paper we present measurements of breakdown voltages in ethanol vapour. In addition, corresponding emission intensity distributions are recorded by the ICCD camera – integrally in visual range of spectra and by using CH filter at 431.2 nm and H_α filter. Measurements are made in a simple parallel-plate electrode system, with a copper cathode [3]. Operating pressures are kept below vapour pressure (~ 40 Torr at room temperature) to avoid formation of liquid droplets.

Paschen curve for ethanol vapour is presented in figure 1. Minimal breakdown voltage of 450 V occurs at relatively low *pd* (0.35 Torr cm), which is typical for electronegative gases. Recorded emission profiles of the discharge revealed the information on the key mechanisms of breakdown. As it is observed, heavy particles have important role in ethanol vapour breakdown. At $E/N > 3$ kTd they become dominant. Most of the emission comes from CH radical excitation, probably created in process of dissociative excitation of ethanol molecule. Our future work includes more detailed spectrally resolved measurements. Time resolved studies will also be performed, to get an insight into kinetics of elementary processes in breakdown.

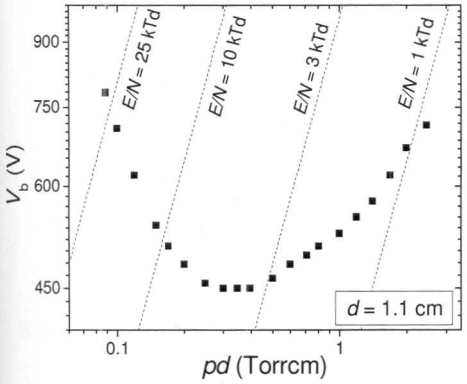

Fig. 1: Paschen curve for ethanol vapour at electrode gap d = 1.1 cm. Several selected values of reduced electric field E/N are indicated by the dashed lines.

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(optical) access to the active plasma is often hampered. On the other hand, time-resolved *in-situ* measurements are a valuable tool (i) to establish temperatures of heavy particles, (ii) to identify excitation mechanisms in the plasma, (iii) to discriminate between gas phase and surface reactions as they occur on significantly different time scales, and (iv) to unravel reaction mechanisms.
This contribution will provide an overview of plasma diagnostic methods to scrutinize the gas phase of dielectric barrier discharges (DBDs) at (sub-)atmospheric pressure conditions thereby covering conventional filamentary and high-current glow-like plasmas. Examples for the implementation of *ex-situ* and *in-situ* methods to laboratory scale reactors as well as to reactors designed for industrial-like processing are provided. Special attention will be paid to time-resolved optical emission spectroscopy that provides a link to electronic excitation channels and hence to the discharge physics. Additionally, infrared absorption spectroscopy using a broadband Fourier-transform spectrometer and modern tuneable semiconductor laser sources will be discussed. Monitoring of molecular species in the mid-infrared spectral range, also known as their "fingerprint region", yields information on the plasma chemistry.
Particularly, time-resolved studies applying pulsed quantum cascade lasers are valuable tools to study the kinetics of plasma-chemical processes and to discriminate gas-phase from surface processes.

IL–18

NEW PHENOMENOLOGY OF GAS BREAKDOWN IN DC AND RF FIELDS

Zoran Lj Petrović, Jelena Sivoš, Marija Savić, Nikola Škoro, Marija Radmilović Rađenović, Gordana Malović and Dragana Marić

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Recent advances in diagnostics and modeling of complex plasma systems opened an opportunity to revisit the breakdown in gases both in DC and RF fields and also in micro gaps. We will first discuss the experimental techniques to determine the breakdown voltage. Typical errors will also be discussed, such as neglecting the long-path breakdown at the left-hand side of the Paschen minimum, together with the electrode conditioning and the measuring properties under an unstable oscillating mode. In addition, we will describe a methodology suitable for establishing the volt-ampere (V-I) characteristics and how to use those with the aim of determining the breakdown voltage.
Time-resolved imaging [1] will be shown describing the development of the anatomy of the discharge in different modes. The spatial profile may be used to establish which of the mechanisms dominate the discharge. The Townsend regime is a low-current diffuse discharge with exponential growth towards the anode [2]. It is necessary to observe such a profile to ascertain that a given discharge operates in the Townsend regime, so that Townsend's theory can be applied to establish the condition for breakdown and the effective secondary electron yield. The temporal development of the normal glow or abnormal glow following the breakdown reveals a transient multi-regime operation that requires a new paradigm.
When considering the V-I characteristics, one may first observe a negative differential resistance in the Townsend regime, which may be explained by a combination of space-charge effects and an energy-dependent secondary electron yield. Thus, the V-I characteristics should be

used in addition to the Paschen curve to determine the secondary electron yields. In what concerns the newly developed field of discharges in and above liquids, we will also analyze breakdown in water vapor and ethanol [3].
RF and microwave breakdowns have a different phenomenology, as the feedback mechanism of secondary-ion production of electrons at the cathode is not necessary. Yet the RF breakdown creates phenomena not often observed in a DC breakdown, such as S-shaped (double valued) Paschen-like curves, frequency/gas number density scaling and additional mechanisms like multipactors. The principal experimental problem with the RF breakdown is the magnitude of the displacement current that thwarts the measurement of the conduction current making it difficult to ascertain the initiation of the discharge.

[1] D Marić, G Malović and Z Lj Petrović Plasma Sources Sci. Technol. 18 (2009) 034009.
[2] Z. Lj. Petrović, et al. IEEE Trans. Plasma Sci. PS 30 (2002) 136-137.
[3] N. Škoro, et al. Phys. Rev. E 84 (2011) 055401(R).

IL–19

PLASMA DEPOSITION OF COMPOSITE MATERIALS WITH METALLIC INCLUSIONS

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Nanocomposite materials based on metallic particles embedded in various carbon-based matrices have been extensively studied during the last decades due to their interesting optical, electrical and biomedical properties. They find an extensive range of applications in sensors, catalytic devices, fuell cells, etc. Among the widely used methods for synthesis of such composite materials are those based on plasma techniques. In this context, this presentation deals with the use of hybrid PVD/PECVD based systems for producing various composite materials with metallic inclusions.
The experimental systems described are equipped with PVD and PECVD plasma sources, mounted perpendicular to one another. The substrate can be either maintained in a fixed position, at 45° with respect to both sources, in the case of a simultaneous exposure to both plasma sources, or can be positioned alternatively in front of each plasma source, when a sequential exposure to each of the plasma sources is chosen.
The PVD plasma source is a magnetron sputtering gun functioning with a metallic target which provides the metallic component of the composite. Depending on the application intended, various targets were used, like Au, Ag, Ni, W, Cu, Ti. Also, several configurations of PECVD plasma sources, from a classical shower-like RF electrode to a plasma jet provided with an injection ring for introduction of the precursors, were designed and implemented in the combined PVD/PECVD set-ups in order to allow a large diversity of materials to be synthesized, from polymeric materials to nanostructured carbon.

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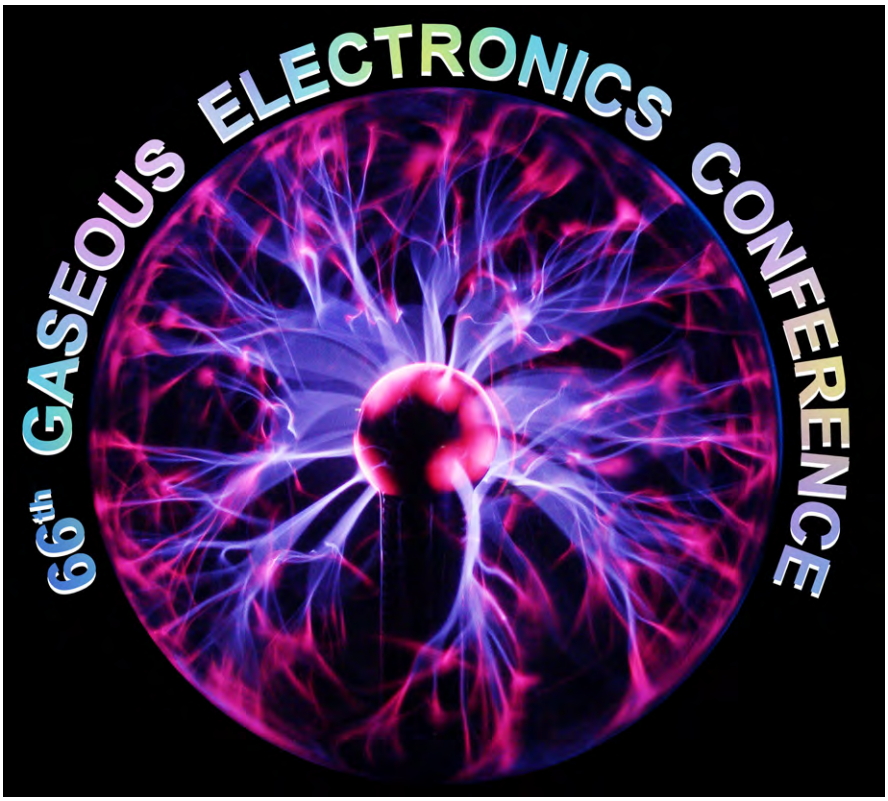

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shock wave, the visible/UV emission spectroscopy was carried out. The arcjet plasma was generated between an anode (copper) and a cathode (Ce/W) with a gap length of 2.5 mm and then expanded through the anode nozzle (throat diameter: 1.0 mm) into low pressure region (expansion section). The discharge current voltage and gas pressure were 40 A ∼30 V and ∼1000 mbar respectively. A visible spectrometer (focal length: 1.0 m, grating: 2400 grooves/mm) was used to measure the plasma emission The electron temperature was evaluated by Boltzmann plot of He I $2p^3Pnd^3D$ series (n =6, 7, 8 and 9), whereas the density was determined by Stark broadening (He I 438.8 nm). It was found that the density significantly increased at the shock region, which can be expected by the simple gas dynamic theory. However, no distinct change of the temperature was observed.

HW1 33 Optogalvanic spectroscopy: Towards a versatile plasma based tool for gas trace analysis* LINA M. HOYOS-CAMPO, A.M. JUAREZ, *Instituto de Ciencias Fisicas, Universidad Nacional Autonoma de Mexico, P.O Box 43-8, Cuernavaca, Morelos, 62251, Mexico* The real-time detection and quantification of molecular traces in atmospheric samples is currently a very active field in medical, homeland security and biological applications. The optogalvanic effect consists in the variation in the electrical properties of a plasma, induced by the interaction of resonant radiation with atoms or molecules present in it. This technique provides a very sensitive and selective spectroscopic tool for gas trace analysis. However, optogalvanic spectroscopy is not currently being exploited thoroughly, in our opinion, in these applications. In the present contribution we will discuss our current efforts towards developing a molecular trace detection facility focused on gas phase volatile compounds (VOC) detection using optogalvanic spectrometry. Our spectrometer consists of a hollow cathode discharge coupled to tunable lasers in the visible (400-800 nm) and mid-infrared, Quantum Cascade Laser (8000 to 10000 nm) spectral range. In particular we will present our preliminary results in the associative ionization induced in a helium (James Lawler, Phys. Rev. A **22**, 3, 1980), as well as an outlook of future work in this emerging area of medical and biological application of gas trace analysis based on optogalvanic spectrometry.

*This work is supported by UNAM program DGAPA-PAPIIT with grant number IT100613.

HW1 34 Particle-In-Cell simulation of a magnetized plasma column exhibiting a non-linear rotating structure JEAN-PIERRE BOEUF, BHASKAR CHAUDHURY, *LAPLACE, CNRS and University of Toulouse, France* STANIMIR KOLEV, *University of Sofia, Bulgaria* A two-dimensional Particle-In-Cell Monte Carlo Collisions (PIC-MCC) model is used to study plasma transport across the magnetic field in a magnetized plasma column sustained by energetic electrons emitted from filaments and injected in the central part of the column. The conditions are similar to those of experimental magnetized plasmas studied for example in the MISTRAL device [1]. Experiments show that the boundary conditions at the end of the plasma column (presence of a limiter, applied voltages) play an essential role in the development of instabilities. Because of the 2D nature of the model, the column is supposed to be uniform in the direction parallel to the magnetic field (only flute instabilities can be described), but electron and ion losses at the ends of the plasma column are taken into account self-consistently in the model. Simulations performed under conditions close to those of the experiments of Ref. [1] (argon, pressure 10^{-2} Pa, magnetic field around 20 mT) predict the formation of a rotating electrostatic plasma structure with spiral arm whose properties are qualitatively and quantitatively close to those observed in the experiments. The model can in particular explain the unexpected distribution of ion velocity measured by Laser Induced Fluorescence in Ref. [1]. We discuss the nature of this instability and its relation with the modified Simon-Hoh instability.

[1] C. Rebont, N. Claire, Th. Pierre, and F. Doveil, Phys. Rev. Lett. **106**, 225006 (2011).

HW1 35 DC breakdown in ethanol vapor* ZORAN PETROVIC, JRELENA SIVOS, NIKOLA SKORO, GORDANA MALOVIC, DRAGANA MARIC, *Institute of Physics University of Belgrade Serbia* DC breakdown is investigated in ethanol vapor at low pressure. Discharge is initiated in parallel-plate electrode system, with copper cathode and transparent conductive anode, 5.4 cm in diameter. The distance between electrodes is adjustable. We present Paschen curve for ethanol measured at electrode separation of 1.1 cm and at pd values between 0.1 Torr cm and 3 Torr cm. Paschen curve has a characteristic shape with a rapid increase of the voltage in the left part and somewhat slower growth in the right-hand branch. The minimum breakdown voltage of 450 V occurs at around pd = 0.35 Torr cm. After breakdown, the discharge operates stable up to pd = 0.7 Torr cm. At higher pd- s, the discharge falls into relaxation oscillations, where it was possible to estimate the breakdown voltages from oscillatory patterns. To investigate elementary processes in the breakdown, for every point of Paschen curve corresponding axial profiles of emission are recorded by ICCD camera. The profiles reveal strong emission peak near the cathode. This indicates that heavy-particle processes play important role in the discharge at all pd values covered by measurements. At the lowest pd values, in the left - hand branch of the Paschen curve, heavy particles (ions, fast atoms and molecules) are dominant.

*Supported by ON 171037 and III 41011 projects

HW1 36 PLASMA BOUNDARIES: SHEATHS, BOUNDARY LAYERS, OTHERS

HW1 37 Characteristics of wall sheath and secondary electron emission under different electron temperature in Hall thruster PING DUAN, HAIJUAN QIN, ANNING CAO, XINWEI ZHOU, *Department of Physics, Dalian Maritime University* LONG CHEN, *School of Physics and Optoelectronic Technology, Dalian University of Technology* HONG GAO, *Department of Physics, Dalian Maritime University* Characteristics of discharge channel wall plasma sheath in Hall thruster have great effects on its performance. In this paper, we establish a two-dimensional physical model in Hall thruster sheath area to investigate the influences of the different electron temperature, propellant and particle weight on sheath potential and secondary electron emission in Hall thruster, by the method of Particle In Cell (PIC) simulation. And the electric field at the particle position is obtained by solving the Poisson's equation. The numerical results show that when the electron temperature is low, the change of sheath potential drop is bigger than that with electrons at high temperature, the surface potential maintains a stable value and the stability of the sheath is good. When the electron temperature is high, the surface potential maintains persistent oscillation, and the stability of the sheath is reduced. Along with the increase of electron temperature, the coefficient of secondary electron emission in wall reduce after the first