

EXPERIMENTAL STUDY OF HIGH PRESSURE GLOW DISCHARGES BASED ON MSE ARRAYS

C. Penache, A. Bräuning-Demian, L. Spielberger and H. Schmidt-Böcking
J.W.Goethe-University Frankfurt, August Euler Str.6, D-60486 Frankfurt am Main, Germany
penache@hsb.uni-frankfurt.de

Abstract

A source of large area, high-pressure glow discharges based on three-dimensional Micro-Structure-Electrode (MSE) arrays is presented. These structures consist of a regular matrix of holes perforated in a thin composite sheet made out of two metallic foils separated by an insulator. Typical dimensions of the presently used MSE are: the holes opening 100 μm , holes pitch 1.5 to 3 mm, electrodes thickness 50 to 100 μm and 50 μm thick dielectric layer. Based on MSE arrays, stable direct current glow discharges were produced in noble gases (Ar, He Ne), air and mixtures thereof over a wide pressure range: hundreds of torr up to one atmosphere. Discharge currents up to 20 mA per hole at moderate forward voltages (around 200 V) were measured. Single hole discharges and systems of up to 200 parallel-operated holes have been studied. The formative time of the discharge, current-voltage characteristics and the emitted radiation were investigated.

1. Introduction

During the past years a lot of efforts were focused towards the generation of stable high-pressure glow discharges due to their potential industrial applications. A number of different approaches (corona and dielectric barrier discharges, direct current, radio frequency and microwave discharges) are known [3-5].

In order to generate large area, dc driven glow discharges at elevated pressures, Micro-Structure-Electrode (MSE) arrays have been implemented [1,2]. For the present investigations we preferred three-dimensional microstructures which are similar with those used in position sensitive gas detectors, known as Gas Electron Multiplier (GEM) [6]. Basically, it consists of a matrix of holes perforated in a multilayer system (metal-insulator-metal). Each hole can be source of a microdischarge that takes place between the two hollow electrodes. The electrode opening, electrode thickness and insulator spacer, have typical dimensions of hundreds of micrometer. The working pressure can be increased up to one atmosphere under moderate voltage conditions (hundreds of volts). The local electric field achieved in the vicinity of the microelectrodes is about 10^5 V/cm, strong enough for plasma generation at atmospheric pressure in noble gases and air.

Electric discharges based on MSE arrays are providing non-thermal plasma. Due to the presence of highly energetic electrons free radicals can be created without heating the gas. Therefore these devices are suitable for gas phase plasma chemistry (reduction of pollutants) and surface processing (cleaning, activation). Moreover, a flexible non-planar design of the MSE allows efficient treatment of peculiar substrate shapes. These discharges are also intense sources of visible and UV radiation and they could be an alternative to conventional light sources (flat panel lamps).

In this paper experimental work is carried out in order to characterise these high-pressure non-equilibrium discharges based on MSE arrays.

2. Experimental setup

The currently used MSE arrays have Cu electrodes of 50 to 100 μm thickness and typically 100 μm holes diameter. The insulator layer, namely 50 μm thick kapton, defines the distance between the electrodes.

In order to make possible the monitoring of individual discharges, the spacing between holes (pitch) was chosen 1.5 or 3 mm. Fig. 1 presents a sketch (cross view) respectively a Raster Electron Microscope (REM) photograph (top view) of a MSE hole. The special profile and the very high precision of the opening are coming from the manufacture procedure.

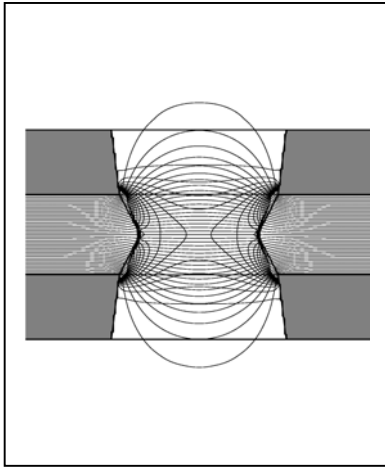


Fig. 1a: Transversal cross-section through a MSE hole and the associated electric field lines.

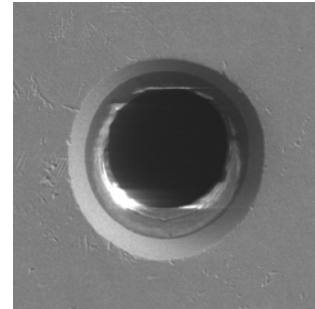


Fig. 1b: REM photograph of a MSE hole. Up charging effects (the white regions) on the kapton surface are present due to the high-energy electron beam used for REM photography.

Single-hole microstructures as well as modules with up to 200 parallel-operated holes have been mounted in a 1000 cm^3 stainless steel vessel. It has two glass/quartz windows to allow the optical access to the MSE sustained discharge. Different gases or gas mixtures are injected in the discharge chamber through a mass flow control system and the working pressure is adjusted with a needle valve placed between the reactor and the pump system.

A load resistor of 40 to 100 $\text{k}\Omega$ is used between the dc power supply (Telenec model TC951) and the powered electrode. The discharge is usually observed end-on at the cathode side with an optical microscope and a digital camera. For spectral measurements the discharge chamber was coupled to a 1 m focal length scanning monochromator (Ortec model BM100). A 1200G/mm grating blazed at 450 nm was used to investigate the emission in the wavelength range 350-850 nm. An Optical Multichannel Analyzer (OMA) with 720 channels assured the detection of the spatially resolved radiation. The dispersion of the system is about 0.2 nm/channel.

3. Electrical and optical investigations

Electric field computation for different MSE systems has been carried out (Software MAFIA 4022). Basic to the simulation is the theory of discrete Maxwell grid equations. An example of static field distribution in a MSE hole is to be seen in Fig. 1a. In the proximity of the microelectrodes the electric field is reaching maximal values up to 10^5 V/cm. Such high values are not necessarily indicating electron field emission as discharge mechanisms but are larger than those required for generation of atmospheric pressure plasma in noble gases or air.

The formative time of breakdown measured in noble gases at low over voltages is about 1 μ s. It indicates that this is not a spark mechanism and it suggests a space charge controlled Townsend discharge (α - γ - σ process).

Electrical characteristics of the MSE sustained discharges, as function of the structure geometry, working gas and pressure, have been studied for single hole discharges and systems of parallel-operated holes. The current-voltage characteristics of a single hole discharge at atmospheric pressure for different working gases are shown in Fig.2a. The U-I characteristic presents different ranges of operation: a Townsend mode (see Fig. 2b), a transition region and a normal glow regime. For low values (less than 0.5 mA) the current increases exponential with the voltage, a signature for a Townsend mechanism. By rising the current, the charge density inside the electrode cavity increases. The space charge at the cathode becomes high enough to distort the axial electric field. A strong radial field develops (cathode fall). The enhanced electric field facilitates ionization processes and correspondingly a fall in sustaining voltage and a rise in current is observed. For further increase in current the voltage drop over the discharge remains relatively constant. An abnormal glow discharge mode was also observed for higher current values.

Fig. 2b presents the electrical characteristic of a 16 holes module operated at 150 torr in Ar+8.5% air. Currents per hole of about 0.1-0.2 mA correspond to the Townsend regime, which exhibits resistive U-I characteristic (positive slope). This makes possible the parallel operation without resistive decoupling [7].

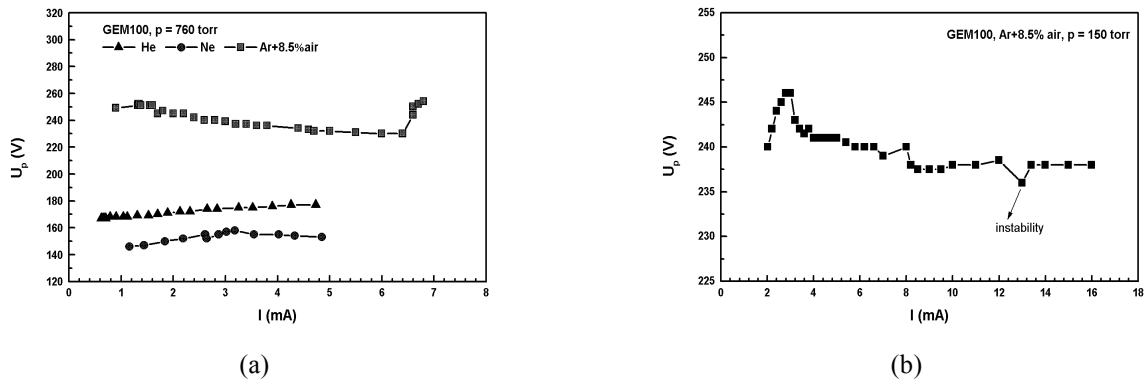


Fig. 2: Voltage-current characteristics of MSE sustained discharges: single hole (a) and 16 holes module (b).

As expected, the optical appearance of the discharge depends on gas pressure and operation mode. For moderate pressures (150-450 torr) it changes according to the U-I characteristic as follow. From a low intensity axial plasma column with diffuse halo to a well defined, very intense column on the axis but not necessarily filling the hole, then the column fills the entire cathode opening and expands out of it.

When the pressure is increased (up to 760 torr) the negatives zones contract. As a result plasma distributes near the cathode walls, the center of the hole remaining dark.

The emission spectra of single Ne and Ar microdischarges have been qualitatively investigated. The light emitted from the cathode region contains the spectrum of the gas as usual for a glow discharge.

In the case of Ar, atomic transitions (ArI) between highly excited electronic states, as well as lines from single charged ions (ArII) are present. In Fig. 3a can be clearly seen that the atomic transitions are dominant (ratio ArII/ArI of about 1/500).

The Ne spectrum indicates only atomic lines, mostly transitions $2p^5 3s^1 - 2p^5 3p^1$. The lack of NeII lines can be explained by the relatively high ionization potential for Ne (21.56 eV) comparing to Ar (15.76 eV).

Metal vapor lines of the cathode material are also present for increased discharge current.

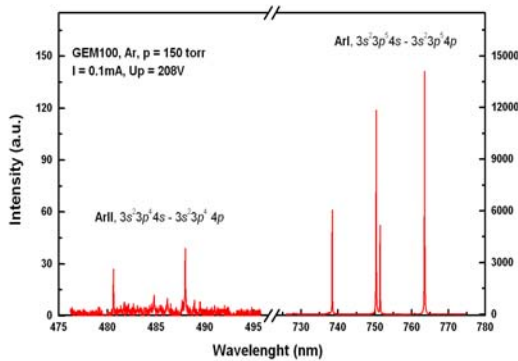


Fig. 3a: Spectra of a microdischarge in Ar at 150 torr and 0.1 mA current. Wavelength range: 476-496 nm and 725-775 nm.

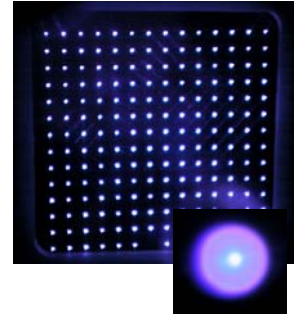


Fig. 3b: Picture of 200 parallel operated discharges at 150 torr without individual resistive ballast. Detailed view of a microdischarge (right down corner).

4. Results and discussion

Based on MSE arrays, stable dc glow discharges were produced over a wide pressure range (150 up to 760 torr) in noble gases (Ar He, Ne), air and mixtures thereof. Parallel operation of up to 200 microdischarges without individual ballast was proved in Ar and Ar/air for pressures up to 150 torr (see Fig. 3b). Systems of 16 or 36 resistive decoupled microdischarges were successfully operated in Ar and Ne at one atmosphere. Current densities from 0,6 to 120 A/cm² respectively power densities of about 0,12 to 24 kW/cm² have been reached.

The very high power density (kW/cm²), especially at the cathode, can lead to constrictions and to glow-to-arc transition. The operation of the MSE arrays in gas flow can help to remove the heat and prevent the onset of such instabilities.

In Ar plasma the thermal effects are much more pronounced than for He and Ne discharges. A possible reason is the high normal current density at the cathode for Ar in respect to Ne or He [7]. The admixture of few percent of air has a stabilizing effect on the Ar discharge, probably due to electron attachment.

Pulsed operation of the MSE arrays can also reduce the thermal stress of the structures. One major disadvantage of the dc driven discharges is the erosion of the cathode. The sputtered metal can deposits on the hole wall and can shorts out the discharge. In order to improve the stability of the MSE, materials as Mo or Ta as electrodes in spite of Cu are desired. Also, due to the low sputtering coefficient and good heat conductivity compared to other insulators, a ceramic substrate (Al₂O₃) is recommended.

A way to get a large area relatively homogenous plasma is increasing the holes density up to 400-500 per cm^2 , which is equivalent with a pitch of about 0.5 mm. Another approach is to use the MSE discharge as "plasma cathode" for a high-pressure glow discharge created between an additional electrode and the microstructure [8]. If the substrate to be treated is metallic can play itself the role of the additional electrode.

Species in the electronic ground state can be characterized by state selective excitation to an excited electronic state and observation of the total fluorescence light (LIF) or via optical absorption spectroscopy (OAS). Measurements of the electron density and temperature of the MSE sustained glow discharge are envisaged.

5. Acknowledgements

The authors like to thank Dr. K. Frank and U. Ernst (Physikalisches Institut 1, Erlangen University) for fruitful discussions and to A. Gandi and R. de Oliveira (CERN) for technical support. This work was supported by: Bundesministerium für Bildung und Forschung (BMBF), Verein Deutsche Ingenieure (VDI), Deutsche Bundesstiftung Umwelt and Land Hessen.

6. References

- [1] M. Roth, T. Haas, M. Lock, K.-H. Gericke, A. Bräuning-Demian, L. Spielberger, H. Schmidt-Böcking, Proc. of 1st Int. Conf. on Microreaction Technology, Springer, Berlin (1998) 62-69.
- [2] P. Scheffler, A. Bräuning-Demian, K.-H. Gericke, C. Geßner, C. Penache, H. Schmidt-Böcking, J. Phys. D, submitted.
- [3] J. Reece Roth, US Pat. No.5,938,854 (1999).
- [4] E. E. Kunhard, K. H Becker, US Pat. No.5,872,426 (1999).
- [5] W. Shi, R. H. Stark, K. H. Schoenbach, IEEE Trans. Plasma Sci. 27, (1999), 16.
- [6] F. Sauli, Nucl. Instr. Meth. A 386 (1996) 531-534.
- [7] Y. P. Raizer, *Gas discharge Physics*, Springer, Berlin, (1997).
- [8] R. H. Schoenbach, R. H. Stark, Appl. Phys. Lett. 74, (1999), 3770-3772.