Formation of Atmospheric Pressure Plasma by Mesh Electrodes and My Desire for its Development

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Since 1987, several research reports about Atmospheric Pressure Glow (APG) plasma in He gas have been published. In 1993 and 1994, an APG-style homogeneous plasma produced by mesh (300-400#) electrodes on ceramic plates in any gas at 50 Hz has been reported, but its stability was only over a 3 mm gap distance. Unpublished Trunec results in 2003 show a time-dependent situation of the plasma-to-filaments discharge. These results open some possibilities of new electrode development along with finding much fundamental knowledge. Namely, two types of discharges can appear in the same apparatus. This discovery might lead to finding new discharge mechanisms after searching through many experimental conditions. Lindmayer's private communication also has surprised me: he could obtain with "other new electrodes" the same results of Lissajous Figures of charges vs voltages as those that were printed in 1993. This shows large possibilities for development of new electrodes which are not mesh, do not show any time dependence and also are more stable over larger gap distances between electrodes.

If any of these new electrodes are developed, surface treatment at atmospheric pressure economically in any gas shall be possible; besides, in the course of such researches, new fundamental knowledge about electric discharges shall appear.

Key words: APG plasma, Metal mesh electrodes, Development of new electrodes.

1. INTRODUCTION

Since 1987 Atmospheric Pressure Glow (APG) plasma in He gas has been developed and applied to many surface treatments: surfaces of large dimension, inner-sides of plastic tubes, powder materials...etc. Fortunately the naming of "APG plasma" in He gas was clearly supported by the long and nice research of Massines-group [1]. They also reported that in extremely pure nitrogen gas the discharge plasma at atmospheric pressure is a Townsend-like (APT) discharge.

The theme in this paper: a homogeneous discharge between dielectric substances with fine metal mesh electrodes of about 300-400 # [2,3], is still not clear. But this plasma shows a glow style even with a 50 Hz source application also in gases such as oxygen, nitrogen, argon and air. It will be a very economical and useful plasma, even better than APG plasma in He, if its stability shall be increased to equal that of APG plasma in He. When this mesh electrodes method was found in 1993, the gap distance of dielectric plates where a stable homogeneous glow is shown was only 3 mm. But



Figure 1. [3] Parallel plate type plasma generator with fine mesh electrodes.



Figure 2. [2] Circuits of current pulse and voltage-charges Lissajou figure.

application of this technique for tetra fluoro ethylene sheet surface treatment by air plasma is already reported[4]. The main researchers about the homogeneous discharge obtained by using metal mesh electrodes are the Trunec and Brablec group (Czech Republic) and the Lindmayer and Tepper group (Germany) since their reports in 1998 [5,6]. Sato (Iwate Univ.) has also started simulation analyses with Trunec since last year [7].

Recently, Trunec's private letter has informed me that the homogeneous discharge will change to the filament style for longer time of discharge; namely, the plasma between ceramic plates with mesh electrodes shows a time-dependent situation. A re-prepared new electrode system shows a beautiful homogeneous plasma also. Then our old results from 1993~1994 and the newest results of the Lindmayer-Tepper group were all corrected with Trunec's information. These results show the continuing importance of the research projects about fine mesh electrodes system or about new electrodes development.

2. BRIEF HISTORY OF FINE MESH ELECTRODES SYSTEM

2.1 First reports [2, 3].

The plasma produced by a mesh electrodes system has been explained in contrast with the APG plasma with He gas, which must use an electric source of over 1 kHz. So its title was written as [Appearance of stable glow discharge in air, argon, oxygen and nitrogen at



Figure 3: [2] (a) An oscillogram of the current pulse of homogeneous plasma. (b) Lissajous figure of voltages-charges of homogenous plasma.



Figure 4. [2] (a) An oscillogram of the current pulse of non homogeneous plasma. A mixture of glow style discharges and filaments in visual observation was written as APG (SED). (b) Lissajous figure of voltages-charges of non homogenous plasma.

atmospheric pressure using a 50 Hz source]. Today, this plasma is not called the APG plasma, because its fundamental nature is not clear.

Along with visual observation of the discharge plasma, an oscillogram of the current pulse and a Lissajous figure of voltage-charges of mesh electrodes system (Figure 1 [3]) were observed using the circuit shown in Figure 2 to confirm whether the discharge state was homogeneous or not. A typical example of homogeneous discharge is shown as APG in Figure 3: (a) current pulse and (b) Lissajous figure. And an example of a mixture of APG plasma and filament style of SED (silent electric discharge, ozonizer discharge) is shown in Figure 4: (a) current pulse, (b) Lissajous figure. Namely, we have measured the current pulse figure to confirm if there really is a homogeneous discharge, based on our experiences of APG plasma of He and Ar-Acetone mixture plasma [8]. In APG plasma, the current shows one pulse / half cycle, so this characteristic phenomenon was used as one confirmation method. But in the plasma between dielectric plates with mesh electrodes, measurements of Lissajous figures of voltage-charges were necessary; in particular, the nitrogen gas plasma situation was not simple.

Examination of the mesh electrodes system application to ozone formation [3] showed that, when pure oxygen is used, energy efficiency was increased by about 20 %, whereas when air is used, energy efficiency was increased by 15 %. We have concluded that increases of molecule-electron collision probability clearly occur. In measurements of ozone formation efficiencies, when nice homogeneity is shown as in Figure 3, the ozone efficiency had a high value; when there was bad homogeneity with filaments as in Figure 4, the ozone efficiency had a low value. 2.2 Studies of the Trunec and Brablec group and the Tepper and Lindmayer group.

First Trunce reported [6] that no disturbance effect caused by the mesh electrodes in electric field occurs beyond a ceramic plate, so he could not explain why mesh electrodes can stabilize the plasma between ceramic-plates. Meanwhile, they continue many farsighted researches on mesh electrodes systems.

Recently, he said in a private letter that he wanted to use a high frequency source to put more energy into the discharge, which is important for the application, e.g. surface treatment. He found that the mesh has no influence at higher frequencies. Nowadays it is also easy to build a power supply with a frequency of several kilohertz. Another problem also involved the mesh electrodes: he needed to use newly prepared electrodes (Al₂O₃ dielectric) because after several hours of APG discharge the process changed to a filament discharge and it was necessary to prepare new (fresh) electrodes. He does not understand this effect.

Trunce's letter about a time dependent-situation of the mesh electrodes system has given me a large shock: if our old data [2, 3] included not real things it would be a large problem. Fortunately, our data were measured always using new electrodes. Also I can find these results in 10-year-old reports: 1. results of one year before could not be repeated; 2. the system that used glass plates with mesh electrodes did not completely change its Lissajous figure in pure oxygen plasma even for long time (about 2 h) discharges.

Tepper's answer to my private question about the time-dependent-situation of the mesh electrodes system was that the nice homogeneous condition could continue for a maximum of 30 minutes in his system of Mayler film dielectrics. And his opinions relating to the stability of the homogeneous discharge are as follows: while the discharge is running, it heats up the discharge vessel. (a) This could have some mechanical impact on the electrode/barrier interface or on the gap distance; (b) the heat could change the properties of the barrier material; (c) the voltage source which drives the discharge could change its voltage amplitude, frequency or its impedance, due to heating up; (d) change of the gas composition could occur.

All these possibilities would have a strong impact on the homogeneity of the discharge. He continued that he can imagine that these parameters are difficult to control over several hours.

He wanted me to stay in contact with Prof. Lindmayer.

2.3 A simulation analysis [7] and private discussion at GD2004 Toulouse last September.

Last year Sato has started analyses for the mesh electrodes system using the data for nitrogen of Trunec [9]. Results were reported using both names to GD2004. Their summary is as follows:

- 1 The field emission effect plays an important role for formation of the non-filamentary atmospheric pressure discharge at 50 Hz.
- 2 The field emission causes a rapid increase of the current and transferred charge, which corresponds to the experimental observation. Without this effect, such time variations of the current and the charge can not be simulated.
- 3 The current pulse calculated has an amplitude of about 1 A and a duration of about 50 ns.
- 4 The calculated electron energy in the discharge is less than 4.5 eV.
- 5 The discharge structure observed at 10 kHz in atmospheric pressure N_2 and He is neither "Townsend-like" nor "glow-like".

Lindmayer has reported also at DG2004 [10]. His work used normal plate electrodes of conducting paste that were painted directly on the barrier. And he examined in detail the surfaces of dielectric materials: Teflon-sprayed glass and $Al_2 O_3$ show a homogeneous gap limit of 3 mm as a maximum value in his experiments at atmospheric pressure.

In a private discussion during GD2004 Toulouse including Lindmayer, Tepper, Sato and Okazaki, Lindmayer has shown some data of Figure 5 [11] which is not yet published. He could obtain a Lissajous figure which is completely the same style as that of Figure 4 (b) [2] for new electrodes which are not mesh.

3. MY DESIRE FOR NEW ELECTRODE DEVELOPMENT FROM CURRENT RESULTS OF RESEARCH ON PLASMA IN MESH ELECTRODES SYSTEM AT ATMOSPHERIC PRESSURE.

Since I found a time-dependent situation of plasma in mesh electrodes, researches on the system have continued. Of course, there may be many works in the world that have not yet been published.

In applications to plasma treatment on many surfaces a time-dependent plasma is not a good plasma. But we can observe homogeneous plasma and filaments in the same apparatus. This experimental condition might give us some very nice breakthroughs to some fundamental research results about atmospheric pressure plasma. Why does homogeneous discharge occur, why are filaments born, in this system can we find some new things experimentally?

Of course, in this research the technique of simulation calculation shall be very useful to analyse such plasmas. Sato has concluded that nitrogen plasma produced using mesh electrodes is not "glow-like" and also not "Townsend-like" [7]. For every gas the fundamental nature of the plasma must be analysed.

This system has relations with many kinds of materials: dielectrics as barrier materials, electroconductive parts as electrodes, gases in mesh style electrodes.....etc. Lindmayer showed a Lissajous figure for a plasma produced with new electrodes but this plasma is a mixture of homogeneous and filaments, as seen in Figure 4. Today we have many kinds of for dielectric materials substances, conductive substances and also intermediate substances. So newelectrode developments have many possibilities. If we can develop a more stable plasma in any gas using new electrodes, and also if it does not change its situation in



Lissajous Figure

Figure 5. [11] Lissajous figure of charges vs voltages for new electrodes which are not mesh.

long-time discharge, the atmospheric plasma application region shall be extended, because this plasma will not use expensive gases such as He.

The fine mesh electrodes only showed the existence of stable homogeneous plasmas in pure Ar, N_2 , O_2 and Air, but not in He at atmospheric pressure even at 50 Hz application; in cases where the plasma does not stabilize at 50 Hz, these gases show more stable plasmas than He does.

And we found in Li's experiment [11] that new electrodes which are not mesh have given the same Lissajous figure of voltage-charges as we found in the old mesh electrodes experiments.

Can we develop new electrodes which can produce a stable homogeneous plasma in any gas?

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