Electrode microwave discharge and plasma self-organization

Yu A Lebedev, I L Epstein, A V Tatarinov and V A Shakhatov
Topchiev Institute of Petrochemical Synthesis RAS, Leninsky Prospect, 29, Moscow, 119991, Russia
lebedev@ips.ac.ru

Abstract. The processes of plasma self-organization and formation of plasma structures at reduced pressures is considered with respect to the electrode microwave (2.45 GHz) discharge (EMD). The term EMD is attributed to discharge maintained near the ending of powered electrode/antenna if plasma region is less than characteristic dimensions of discharge chamber. EMD is generated at gas pressures ranged between 0.5 and 400 Torr, plasma gases are Ne, Ar, H₂, N₂, O₂, air and mixtures containing CH₄ and C₂H₂. Electrodes/antennas of different shapes were used to ignite the discharge: solid and tube cylindrical antennas with diameter of 1-6 mm, needle, spiral and trident antennas etc. Plasma absorbed power is less than 30 W. Phenomenology of EMD is presented. Results of emission spectroscopy and probe measurements together with results of EMD modelling are described. The concept of “self-sustained-non-self-sustained” mechanism of EMD is concluded from analysis of publications on microwave discharges and results of study of EMD. Areas of possible EMD application are discussed.

1. Introduction
A lot of publications are now devoted to the processes of plasma self-organization and to formation of plasma structures at reduced pressures [1-10]. These phenomena are clearly appeared in the electrode microwave discharge (EMD). Here we call as EMD the discharge when plasma is ignited near the tip of powered electrode/antenna and luminous plasma region is less than dimensions of discharge chamber. Discharge turns into the ordinary discharge mode [11] at smaller distance between the electrodes.

Interest to the electrode microwave discharge is caused by its distinctive features:
- Extremely low maintaining discharge power (1-2 W),
- Wide range of operation pressures,
- Generation of compact plasma structures,
- Generation of plasma active particles in large vessels,
- Generation of plasma in the chosen point of space (localization of plasma region),
- Generation of plasma active particles near the treated surface,
- Absence of pollution of plasma treated surfaces by the products of electrode erosion,
- The area of plasma region is controlled by the electrode shape.

Electrode microwave discharge gives the possibility: to study and realize the plasma chemical processes, to intensify physical-chemical processes in gas phase, including the process of burning, to study the interaction of plasma with solids, to study the behavior of solids surrounded by plasma with gas flows, to study the interaction of plasma clots with gas flows.
Intensive study of EMD was started in 1997 and now we have a lot of knowledge on discharge physics and on application of the discharge.

2. Experimental set-up

Detailed description of experimental set-up was presented in [12-18]. Stainless steel cylindrical discharge chambers with diameters of discharge vessel of $R_1=7$ cm and $R_2=4.2$ cm were used for experiments. Plasmas in both arrangements revealed similar features, thus the properties of the described plasmas seem to be typical for the electrode microwave discharge systems. Plasma gases were Ar, Ne, $H_2$, $N_2$, $O_2$, CH$_4$, C$_2$H$_2$, air, and their mixtures at pressures 0.5-400 Torr. Gas flow systems were used providing the total gas flow rate less than 1000 sccm. System “gas feeding-pumping” provides possibility to operate with gas mixtures of 3 gases. The latest modification of setup (stand EMD-3) is shown in figures 1, 2.

![Figure 1. Schema of experimental setup EMD-3](image1)

![Figure 2. Vacuum system of EMD-3](image2)

A tubular and solid stainless steel and copper electrodes of different shapes (direct cylinder and bent electrodes, trident electrode, spiral electrode, etc.) with diameters of 0.5-6 mm have been used as the antennas.

Most of results have been obtained when the discharge was ignited at the end of cylindrical electrode (antenna). Microwave power (2.45 GHz) was transmitted from the magnetron generators with output powers of 2.5 kW or 150 W. The power absorbed in the system was measured by a directional coupler. The power absorbed in the discharge was obtained by subtracting of power losses in chamber without plasma from the power losses with plasma at the same incident power.

The set-up was equipped with the system of optical measurements (monochromators MDR-4 and MDR-23 with photo registration, spectrograph AvaSpec-2048). Discharge visualization was made with video camera (exposition times were ranged between 1/20 and 1/8000 s) or with digital photo camera. Some results are shown in figures 3, 4.

Methods used for experimental study:

- Photometry of the discharge (study of EMD structure).
- Double probe method (determination of local values of electron temperature and density) [14-23].
- Emission spectroscopy in visible range of spectrum with spatial resolution (determination of plasma particles concentrations, gas temperature and electric fields, etc) [17,18,21-23,25-29].
- Electrodynamic measurements (determination of plasma absorbed power).

Methods of EMD simulation:
• Self-consistent one dimension modelling in quasi-static approximation [24]
• Self-consistent 2D modelling on the base of Maxwell equation [30,31,33]

Figure 3. View of the air discharge from the tip of 6 mm electrode at different time of exposition (2 Torr)

Figure 4. View of the hydrogen discharge with needle (1 mm) electrode at different time of exposition (47 Torr)

3. General features and peculiarities of EMD in molecular gases and mixtures with noble gases

I. Discharge properties are independent of diameter of the chamber.
II. Discharge consists of:
   - Bright thin (1-3mm) near electrode plasma region which covers the tip of the electrode
   - Ball-like plasma region
   - Dark external space
III. Ball dimension increases with microwave power and decreases with pressure.
IV. Upper limit of microwave power exists. It is impossible to keep the plasma region at the tip of electrode at higher power (discharge runs away along the antenna towards the microwave generator).
IV. At high microwave power and pressure the electrode can be destroyed because of melting and even explosion when only thin near electrode plasma exists.

Plasma ball has sharp external boundary. Probe measurements showed an abrupt decrease of electron concentration in thin boundary layer even in diffusion controlled regime (see below). This boundary reveals the elastic property being in contact with external body, e.g. with the probe. In the case of coexistence of two close placed balls (near two electrode/antenna) increase in microwave power first lead to increase of ball diameter. Further increase of the power brings to ball deformation into ellipse and dark space always exists between two plasma regions.

4. Some results of experimental study

4.1. Electrode erosion. It was indicated that the risk of contamination of plasma vessel and treated surfaces in EMD by products of electrode erosion is small [12,13]. In experiments on the diamond film deposition on a silicon substrate in a microwave electrode-discharge plasma under fairly severe conditions (the electrode was heated to the red-heat temperature, and the temperature of the substrate located at a distance of 1 cm from it was 1000 °C), ESCA analysis showed no traces of the products of electrode erosion in plasma-treated substrates. This is distinctive feature of microwave plasma.

This problem has been studied intensively for RF and DC discharges. As the field frequency increases, the thickness of the electrode sheath decreases (the electron density is high enough and
exceeds the critical density) and the energy of the ions bombarding the electrode also decreases. In the microwave range, the sheath conductivity is low, and the current continuity is provided by the displacement current through the capacity of the sheath. In this case, the role of the electrode in generation of charged particles is negligible (γ-processes associated with the secondary electron emission from the electrodes can be neglected), and the discharge exists in the form of an α-discharge, in which the processes of volume ionization are of importance. In this case the ion bombardment of the electrode is similar to that for discharge wall in glow discharge.

4.2. Spectroscopy of EMD. Distributions of integral over the spectrum emission of EMD plasma showed strongly nonuniform structure of the discharge depending on pressure and microwave power (Figure 5, 6) [17,18]. Similar behavior has the emissions of plasma lines and bands. Total emission of EMD showed close to linear dependence against microwave power. Emission of the discharge is concentrated in near electrode region. Thus the total emission of EMD characterizes the brightness part of the discharge and can be used to define it parameters.

![Figure 5](image1.png)  
**Figure 5.** Axial distribution of integral EMD emission in hydrogen

![Figure 6](image2.png)  
**Figure 6.** Axial distribution of integral EMD emission in nitrogen

Gas temperature in hydrogen EMD plasma was defined by the relative intensities of rotational lines of the electron excited molecules H\textsubscript{2}(d\textsuperscript{2}Π\textsubscript{u}). Calculations were based on intensities of Q and R-branches emission for diagonal (v'=v''=0,1,2) bands of Fulcher α-system H\textsubscript{2}(d\textsuperscript{2}Π\textsubscript{u}→a\textsuperscript{3}Σ\textsubscript{g}). Rotational temperature of the ground state of H\textsubscript{2} was calculated taking into account the ratio of rotational constants of the ground and excited states. The temperature had flat radial distribution and does not exceed 700 K at pressures below 10 Torr [27].

Method of analysis of non-resolved spectra of the 1\textsuperscript{st} negative and 2\textsuperscript{nd} positive system of nitrogen emission was used to define rotational temperature of nitrogen plasma. At known conditions this temperature can be interpreted as equal to the gas temperature. Defined gas temperature was less than 600 K. Heat conductivity equation also was used for calculation of gas temperature.

Degree of hydrogen dissociation was defined by actinometry method [20]. It was shown that degree of dissociation is less than 1%.

Method of related intensities of lines emission was modified to the conditions of electrode hydrogen microwave discharge to define the electron density and electric field strength profiles [28]. Light emission of H\textsubscript{α}, H\textsubscript{β} and Ar lines were used. Homogeneous Boltzmann equation was used to calculate the excitation coefficients under the direct electron impact. In the case of direct electron impact excitation both hydrogen and argon emission simple formulas define the relations of ratios of line intensities with the electron density and electric field strength. The main result is that electron density in bright near electrode sheath exceeds the critical density, but in the plasma ball the density is under critical.
In the case of nitrogen plasma the problem to define the electric field strength and electron density was also solved but the procedure is much more complicated [32]. The method is based on comparison of measured and simulated spectra of
\[ N_2(C^1\Pi_u \rightarrow B^3\Pi_g) \text{ nitrogen emission.} \]
The simulated spectrum is a result of simultaneous solution of balance equations for excited particles and the Boltzmann equation for electron energy distribution function (EEDF). Following particles were taken into account: nitrogen molecules in the ground \( X^1\Sigma^+ \) state (47 vibrational levels, the vibrational level \( v = 46 \) being taken as the level of dissociation of the nitrogen molecule via vibrational excitation) and in the electronically excited \( A^3\Sigma^+_u, B^3\Pi_g, W^3\Delta_u, B^3\Sigma^-_u, C^3\Pi_u \) (5 vibrational levels), \( E^3\Sigma^+_g, D^3\Sigma^+_g, a^1\Sigma^-_u, a^1\Pi_g, w^1\Delta_u, a'^1\Sigma^+_u \) states; nitrogen atoms in the ground \(^1S\) state and excited \(^2P\) and \(^2D\) states; and electrons \( e \).

The system of equations was solved numerically. At the initial moment of time distribution function on vibrational and rotational energy levels of molecules in the ground state corresponds to Boltzmann distribution at value of the vibrational temperature equal to the gas temperature. During integration of the balance equations rate constants of the vibrational excitation were recalculated depending on the vibrational temperature of the first vibrational level (300 K ≤ \( T_v \) ≤ 6000 K).

The method was verified by comparison of calculated results with known data of rate coefficients for excitation of metastable \( A^3\Sigma^+_u \) state and electron excited \( C^3\Pi_u \) state of nitrogen molecule, plasma chemical composition, vibrational temperature \( T_v \), EEDF and its the basic moments, such as Townsend’s coefficient \( \alpha/N \), the drift velocity \( v_{dr} \), the electron temperature \( T_e \) and characteristic energy \( D/\mu \). It was also checked by comparison of calculated and measured values of electric field strength and plasma density in DC glow. The calculated and known results were in good agreement.

The method was used for diagnostics of EMD and showed that in nitrogen plasma just as in the hydrogen plasma, electron density in near electrode region was higher than critical one whereas in the spherical part it was less than critical value. In conventional microwave discharges overcritical plasma was observed in the self-sustained discharges and under-critical plasma is a characteristic feature of non-self-sustained discharges.

4.3. **Probe diagnostics of EMD.** Double probe technique was used for determination of properties of charged component of EMD plasma.

4.3.1. **Properties of luminous part of EMD.** Some results on parameters of nitrogen plasma electrons and distributions of DC voltage in the plasma are presented in Figure 7 [18,19].

![Figure 7](image_url)
It is seen that the electron temperature monotonously decreases with radius whereas the ion
saturation current decreases abruptly at the boundary. Measurements showed also that the voltage-
current characteristics of double probe system passes through the zero current at a certain DC voltage
difference between the probes $\Delta U_p(I_p=0)$. The last one falls down to zero in the external region of the
discharge.

Some comment can be done on the nature of $\Delta U_p(I_p=0)$. There are several sources of the DC
voltage. The first one is the well-known rectification effect of microwaves on non-linearity of the
probe-to-plasma sheath. In the case of strong non-homogeneous plasma differences in amplitudes of
the microwave field strength in the points where the probes were placed leads $\Delta U_p(I_p=0)\neq 0$. This
source can be considered as a disturbing factor for probe measurements. We have used the
resistive filter to suppress the influence of ac field, which was successfully used in microwave plasma
measurements. The indirect evidences should be taken into account. Analysis fulfilled in [29] gives the
evidence that the DC voltage really exists in the plasma. It can be related with nonlinear interaction of
electromagnetic waves with the overcritical plasma accompanied by frequency transformation.

Observed rapid decrease of electron density at the boundary of plasma ball is very important and
interesting result in the light of creation of plasma structure. It should be noted that the experimental
conditions corresponding to figure 7 can be attributed to diffusion controlled regime. The role of
processes of volume losses of charged particles such as volume recombination and attachment is
negligible. This means that the only reason to suppress the spread of charged particles to the external
wall is existence of local electrostatic field. Such a property has the double layer. It is known that an
intermediate double layer with strong charge separation and appropriate electrostatic filed exists in
discharges with abrupt jump of overall dimensions or between two quasi-neutral plasmas come into
contact. The reason why the double layer is appeared in a fixed point of space depending on pressure
and power is still under consideration. It is clear that this plasma property defines the process of self-
organization.

4.3.2. *Properties of dark part of EMD*. It was shown that the dark space surrounding the plasma ball
contains charged particles [14-16] (see figure 8). The space outside the plasma region is spatially
uniform and double probe voltage-current characteristics pass through zero current at $\Delta U_p(I_p=0)=0$.

Estimations have shown that the characteristic length of electron energy losses in collisions with
heavy particles is much less than the observed length. This means that figure 8 illustrates the heating
effect of the electromagnetic field. Thus the electric field strength distribution can be determined with
the help of the Boltzmann equation and measured values of electron temperatures. It is possible to
match the electric field strength in such a way that the solution of the Boltzmann equation gives the
electron energy distribution function with the mean electron energy corresponding to the measured
value.
5. Recent results of EMD modelling

2D model designed in [30,31] on the base of combined solution of non-stationary Maxwell equations and balance equations for charged particles for quasi-neutral plasma was modified for the geometry of EMD [33]. All necessary properties of electron component of plasma were obtained by numerical solution of homogeneous Boltzmann equation. It was used for analysis of some properties of EMD hydrogen discharge.
5.1. Plasma density profile. Calculations showed that maximum of plasma density always placed near the tip of the electrode at the distance of 1 mm from it surface. The maximum value exceeds the critical density \( (7 \times 10^{10} \text{ cm}^{-3}) \) with the factor 2-5. At low pressure (0.5 -1 Torr) when plasma resonance is pronounced, the field after the rapid fall down near the electrode have the maximum at the point of plasma resonance at some distance from the electrode. After this point the plasma density corresponds to diffusion profile. At higher pressures (2-8 Torr) the role of resonance is small and diffusion plasma density profile exists in wider region.

5.2. Matching of the generator with chamber filled with plasma. It is known that the form of discharge when plasma exists on the edge of the antenna in such a system is not the only form of the discharge. It is possible to produce several discharge zones along the antenna or the discharge is concentrated near the window at the entrance of antenna into the chamber. All these discharges are the electrode discharges but the object of our study is the discharge at the tip of the electrode only. Thus the problem of matching is considered regarding this type of discharge.

Calculations showed that at pressures below 2 Torr close to full matching was achieved at the length of antenna \( L_{el} = n\lambda \), where \( \lambda \) is the wavelength in free space, \( n \) is the integer. At higher pressures the best matching was observed at \( L_{el} = \lambda(n + 1/4) \). These results became clear if one will take into account that at small pressures plasma disturbs the field due to plasma resonance (plasma impedance has large reactive component). At higher pressures the resonance is absent and plasma impedance is active.

5.3. Plasma region dimension. The problem considered above is related with problem of determination of maximum absorbed power at which the discharge exists at the tip of the electrode. This problem can be solved only on the base of solution of Maxwell equations. Figures 10 illustrate dependence of discharge structure on microwave power and transition from discharge on the tip of electrode to two discharges (near the entrance in the chamber and at the tip of the electrode).

It was shown that electrodynamics of discharge chamber defines the maximal power consumption and hence the maximum size of plasma.

At small incident powers the discharge is initiated at the tip of antenna where electric field strength is several times increased. The size of plasma region increases with increasing incident power and discharge begins to spread towards the generator along the antenna, but its center is still fixed at the tip of antenna. The increased size of plasma shields the field penetration to the tip. The role of this point in the discharge position stabilization is decreased. At definite conditions the discharge is detached from the tip of antenna and moves towards the generator. At the same time the electric field strength at the entrance in the chamber also increases with increased incident power (the diameter of outer electrode in this zone is much less than in the chamber and the electric field strength is higher). The probability to ignite discharge in this region increases. In this case this region absorbs the main part of power.

Thus the upper level of power exists which can be absorbed in the discharge at the tip of the electrode. This power defines the largest size of plasma region. The largest size of plasma is defined by the field frequency, other thing being equal.

6. Application of EMD
In spite of short history of EMD study now is clear that this discharge can be effectively used in different plasma based processes. It has already published that it can be applied for:

- Diamond deposition [12,13,34].
- Deposition of CNx films and nanotubes [35-37].
- Depositions of metal films (Cu, Al) [37].
- Deposits of TiN и TiO2 films [37].

Some known EMD disadvantages should be taken into account for technological applications:
- Considerable non-uniformity of plasma – low spatial homogeneity of deposited films.
- Droplet like structure of metal, metal oxide and nitride films was observed.
- Low upper level of plasma absorbed power.

![Figure 10](image)

**Figure 10.** Results of 2D self-consistent modelling of hydrogen plasma at incident power 30 W (a), 70 W (b), and 150 w (c). Pictures on the top side show electron density distributions and the bottom side – distributions of electromagnetic field.

### 7. Conclusion on physics of EMD

Analysis of publications on microwave discharges, experimental results and results of modeling of electrode microwave discharge are the base to formulate and substantiate the assumption on “self-sustained-non-self-sustained” mechanism of the discharge. The discharge consists of the region of self-sustained discharge (near electrode region with overcritical plasma density) which is surrounded by the region of non-self-sustained discharge (ball shaped region with under critical plasma density). The layer with charge separation (the type of the double layer) exists on the outer discharge boundary which provides the abrupt decrease of plasma density. Creation of this layer leads to plasma self-organization.

### Acknowledgments

This study was supported by RFBR Grant 02-02-16021, Project #20 of the Program of fundamental research of the Presidium of the Russian Academy of Sciences and NWO-RFBR grant 047.016.019.

### References

[1] Nerush O A, Novopashin S A, Radchenko V V, Sukhinin G I 1997 Spherical strata in 3-D Glow discharge *Preprint* N 285-97 Inst. of Thermophysics, Siberian Branch RAS
[2] Nerush O A, Novopashin S A, Radchenko V V, Sukhinin G I 1998 Phys. Rev. E 58 4897
[3] Conde L, Leon L 1999 IEEE Trans. PS 27 80
[4] Ivanov S T, Thomae R W, Klein H, Hilschert F H, Nikolaev N I 1998 *Bulgarian Journal of Physics* 25 49
[5] Gildenburg V B , Markov G A 1982 *Pis’ma v ZhTF* (in Russian) 8 1245
[6] Terebessy T, Kudela J, Kando M 2001 *Microwave Discharges: Fundamentals and*
Applications. Ed. Yu.A. Lebedev (Yanus-K, Moscow), pp 163-74.

[7] Kirichenko A Ya, Martyniuk S P, Motornenko A P, Skuratovsky I G, Suvorova O A 2002 Pis'ma v ZhTF (in Russian) 28 164

[8] Batanov G M, Berezhetskaya N K, Bol'shakov E F et al. 1993 Plasma Sources Sci.&Technol 2 164

[9] Brovkin V G, Kolesnichenko Yu F, Khmara D V 1994 Ball Lightning in the Laboratory, eds. R F Avramenko et al (Moscow: Chemistry) p 119

[10] Brovkin V G, Kolesnichenko Yu F 1994 Tech. Phys 39 222

[11] Brandt A A, Tikhomirov Yu V 1974 Plasma multipliers of frequency (Moscow, Nauka, in Russian)

[12] Bardos L, Barankova H, Lebedev Yu A, Berg S, Nyberg T 1996 Proc. European Conf. on Diamond, Diamond-like and Related Materials p 4.1.

[13] Bardos L, Barankova H, Lebedev Yu A, Nyberg T, Berg S 1997 Diamond and Related Materials 6 224

[14] Bardos L, Lebedev Yu A 1998 Technical Physics 43 1428

[15] Lebedev Yu A 1998 Proc. Russian Conf. on Physics of Low Temperature Plasma (Petrozavodsk) p 254

[16] Bardos L, Lebedev Yu A 1998 Plasma Phys. Reports 24 956

[17] Lebedev Yu A, Mokeev M V, Tatarinov A V 2000 Plasma Phys. Reports 26 272

[18] Lebedev Yu A, Mokeev M V 2000 High Temp. 38 338

[19] Bardos L, Lebedev Yu A 2000 High Temp. 38 528

[20] Bardos L, Lebedev Yu A 2001 Plasma Phys Reports 27 418

[21] Lebedev Yu A, Mokeev M V, Tatarinov A V 2000 Proc. Int. Workshop Strong Microwaves in Plasmas (N-Novgorod) vol.1 (Institute of Applied Physics RAS) p 271

[22] Lebedev Yu A, Mokeev M V, Tatarinov A V, Epstein I L 2001 Proc. IV Int. Workshop “Microwave Discharges: Fundamentals and Applications” Zvenigorod, Russia (Moscow, Publ. Comp.”Yanus-K”) p.127

[23] Lebedev Yu A, Mokeev M V 2002 Technical Physics 47 135

[24] Lebedev Yu A, Tatarinov A V, Epstein I L 2002 Plasma Sources Sci & Technol. 11 146

[25] Lebedev Yu A, Mokeev M V, Tatarinov A V, Epstein I L Ed. By A.G. Litvak. 2003 Proc. Int. Workshop Strong Microwaves in Plasmas (Nizhny Novgorod) (Inst. of Applied Physics RAS) p 714

[26] Lebedev Yu A, Mokeev M V, Tatarinov A V 2003 Proc. IV Int. Workshop Microwave Discharges: Fundamentals and Application (Greifswald, Germany) (INP) p 55

[27] Lebedev Yu A, Mokeev M V 2003 Plasma Phys. Reports 29 251

[28] Lebedev Yu A, Mokeev M 2003 Plasma Phys. Reports 29 983

[29] Lebedev Yu A, Mokeev M 2003 High Temp. 41 725

[30] Lebedev Yu A, Tatarinov A 2004 Plasma Sources: Sci&Technol. 13 1

[31] Lebedev Yu A, Mokeev M V, Tatarinov A V, Epstein I L 2004 Plasma Phys. Reports 30 91

[32] Lebedev Yu A, Shakhatov V A 2005 Plasma Phys. Reports (in press)

[33] Lebedev Yu A, Tatarinov A V, Epstein I L 2006 High Temp (in press)

[34] Taniyama N, Kudo M, Matsumoto O, Kawarada H 2001 Jpn. J. Appl. Phys 40 L698

[35] Bardos L, Barankova H, Lebedev Yu A 42-nd Ann. Conf. of Soc. of Vac. Coaters, Chicago, IL, April 17-22, Proc. SVC TC (1999), paper E-7

[36] Nagasaka M, Kando M 2000 Proc. 17th Symp on Plasma Processing (Japan) p 579

[37] Eto A, Kimura S, Kando M 2003 Proc. 5 Int. Workshop Microwave Discharges: Fundamentals and Application (Greifswald) p15