UNIPOLAR AND ELECTRODELESS CAPACITIVELY COUPLED HIGH-FREQUENCY DISCHARGES EXCITED AT ATMOSPEHRIC PRESSURE AND THEIR APPLICATIONS*

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УНИПОЛЯРНЫЙ И БЕЗЭЛЕКТРОДНЫЙ ЁМКОСТНО СВЯЗАННЫЙ ВЫСОКОЧАСТОТНЫЙ РАЗРЯДЫ ПРИ АТМОСФЕРНОМ ДАВЛЕНИИ И ИХ ПРИМЕНЕНИЯ

The set of experimental results proves the unknown fact that the torch discharge excited in molecular gases acts as an effective atomizer. This creates ideal conditions for same plasmachemical reactions and for the application in spectral analysis.

The apparatus for the generation of discharges of a new type is described; the stable electrodeless capacitively coupled highfrequency discharge and obstructed high-frequency discharge.

The torch discharge burning in molecular gases acts as an effective atomizer. This fact provides ideal conditions for some plasmochemical reactions. The torch discharge and torch arc are advantageous to be applied in spectral analysis.

Similarly, the stable electrodeless capacitively coupled high frequency discharge and obstructed high frequency discharge consisting of an eleminated electrode layer only may be employed.

Unipolar high frequency (further abbreviated hf) discharges burn on electrodes with hf voltage about 2 kV at a generator frequency of 4 MHz or more, at atmospheric pressure. According to power output liberated in the discharge we may differentiate between the hf corona, the torch discharge and the torch arc [1].

DISSOCIATIVE RECOMBINATION PROCESSES IN A TORCH DISCHARGE

In molecular gases the dissociative recombination plays an important role during the energy transfer into the torch discharge plasma. It is spectrally proved that positive ions are always present within this discharge electrode layer. The positive ions space charge produces an electric field which induces the Fowler-Nordheim electrode electron emission [2]. With a strong electric field the electrons in the electrode layer obtain a considerably increased kinetic energy, which is transferred at excitation or ionization in elastic collisions to atoms or molecules of the torch discharge plasma.

In the torch discharge burning in air or nitrogen at atmospheric pressure there appear in the electrode layer molecular ions N_2^+ having the excitation potential 18 eV. These molecular ions diffuse from the electrode layer into the torch discharge plasma, where due to collisions with electrons they decay by dissociative recombination into atomic nitrogen [3]. Since the molecular nitrogen dissociative energy is 9.7 eV, the originating atoms have the kinetic energy 8.3 eV, which, in collision with the nearest molecules, dissipates rapidly into a predominantly chaotic thermic motion. In a torch discharge, which burns in molecular gases, only a small part of energy becomes lost as radiative energy [4].

This process of the energy transport into the torch discharge plasma by means of a dissociative recombination not only proves the accordance between the calculated and the measured energy of the plasma volume unit, but it also shows the temperature of discharges in molecular gases with an increased dissociative energy to be relatively enhanced if compared with that of discharges burning in molecular gases having a reduced dissociative energy [3]. At the association of dissociated molecules the greater energy is liberated in the torch discharge plasma, when the increased is the dissociative energy of molecular gases in which the discharge burns.

An important result if the investigation appears to be the knowledge that the torch discharge burning in molecular gases produces a considerable atomization of the applied gases.

To verify the above fact the parabolic shape of a horizontally buring torch discharge flame is investigated. The values of 15 % to 25 % of the atomization obtained for the discharge burning at atmospheric pressure [5] have been discussed in [6].

Yellow-green chemoluminiscent radiation observed at the point of the torch discharge burning in air, especial under greater power outputs, may satisfactorily be expressed by reactions requiring the presence of atomic oxygen

$$O + NO_2 = NO + O_2 .$$

This reaction is followed by another producing the chemoluminiscent radiation just mentioned

$$O + NO = NO_2 + h\nu .$$

Since the latter reaction course is much slower than the former one, the entire in the discharge existing NO_2 is converted into NO. On analyzing the torch discharge the presence of NO_2 in air has not been established [7], but an increased NO volume has been found [8].

PLASMOCHEMICAL REACTIONS IN TORCH DISCHARGE

In the torch discharge the hitherto investigated plasmochemical reactions proceed as follows: a) occurrence of NO from air and admixture of O_2 with N_2 [8], b) occurrence of C_2H_2 from CH_4 and admixture of CH_4 with H_2 [9], c) occurrence of O_3 form O_2 and air [9].

The yields of all the given reactions are much higher than we may obtain by calculation of the thermodynamical processes under the most convenient conditions [10]. The great values mentioned for the plasmochemical reactions are, however, comprehensible enough, if the reaction to take place between the atomized molecules are taken into account. In this case the thermodynamic of open non-equilibrium systems must be considered.

The above information on the atomization of molecular gases in the torch discharge has successfully been applied to the conversion of $SiCl_4$ into $SiHCl_3$ in hydrogen [11].

UNIPOLAR HIGH FREQUENCY DISCHARGES AS SOURCES OF SPECTRA EXCITATION IN SPECTRAL ANALYSIS

Torch discharge plasma is formed by gas in which this discharge burns [1], this is the reason why this discharge is appropriate for gas analysis.

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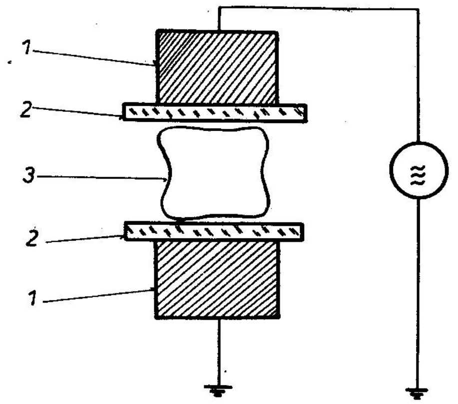
Most studies employing the torch discharge for spectral analysis have proceeded so that the analyzed solution aerosol is introduced into the torch discharge.

The torch arc creates plasma from vapour of material of the electrode on which it burns [1], therefore, it may well be used for spectral analysis of metals and semiconductors of which the electrode is made [12].



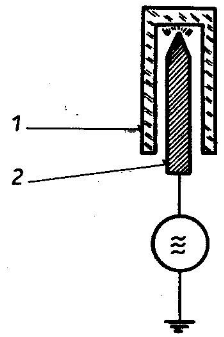


Fig. 1. The electrode configuration of the obstructed high frequency discharge. 1 — sillica tube with sealed bottom, 2 — metal electrode.

Fig. 2. Arrangement for generation of stable electrodeless capacitively coupled high frequency discharge. 1 — metal electrode, 2 — dielectric plate, 3 — discharge.

A very fast method is developed in the spectral analysis of minerals and ores where, without any modification, the above samples may be inserted into the torch discharge flame directly. The torch discharge plasma glows the sample and the discharge flame above the sample becomes spectrally coloured. In the case of some conducting materials the sample produces an electrode layer in which a strong sputtering of the analyzed material, due to electron bombardment, takes place.

The temperature of the electrode which burns on the torch discharge is relatively low, therefore the analysis of some elements (e. g. Ca, B) on the carbon electrode cannot be suppressed because of the occurrence of carbides.

Most spectral lines are found in the electrode layer on the torch discharge or the torch arc. This electrode layer ignition may initiate on the electrode even without any additive components of these discharges.

OBSTRUCTED HIGH FREQUENCY DISCHARGE, ELIMINATION OF ELECTRODE LAYER

A pointed metal or carbon electrode of diameter 2 to 3 mm is inserted into a closely fitting quartz tube sealed on both ends (Fig. 1). If we supply a hf voltage of 2 to 3 kV to this electrode it originates by self-ignition an electrode layer on the electrode point inside the quartz tube. The sealed end of the tube above this electrode point prevents the torch discharge to occur. If we increase the hf voltage supply the torch discharge ignition may appear even above the discharge tube. Namely the current supplied through the capacity between the electrode and the discharge above the sealed end of the quartz tube is strong enough to discharge generation [12].

ELECTRODELESS STABLE CAPACITIVELY COUPLED HIGH FREQUENCY DISCHARGE AT ATMOSPHERIC PRESSURE

Electrodeless capacitively coupled hf discharge burning at atmospheric and/or related pressure has hitherto been generated in a cylindrical discharge tube rounded by two metal electrodes separated from each other supplied by hf voltage. In such a geometrical configuration the discharge space closes the scattered lines of force only and the originated constricted discharges [13] due to very unstable and unceasingly varying their position [14].

A homogeneous flow of lines of force has a geometrical arrangement which is constructed of two metal electrodes whose endsare plane surfaces. These electrodes are placed opposite each other so that the plane surface are mutually parallel with the discharge space between. On the plane surface of each electrode there is superposed a dielectric plate bounded by mutually parallel plane surfaces again, Fig. 2. These dielectric desks make the direct connection of the discharge plasma with the metal electrodes impossible and provide the capacity hf current for the discharge. In the discharge space the stable hf discharge ignition may be realized in air under atmospheric pressure if we supply the electrodes with hf voltage of 2 to 3 kV. Such a discharge may originate in argon at the dielectric plates at a distance about 8 mm by selfignition; in air at atmospheric pressure the igniting of the discharge is made easier by inserting a sharp metal point into the discharge space.

Using the stable capacitively coupled hf electrodeless discharge generated in the above way may be of advantage for some plasmochemical reactions, where the metal electrode is inapplicable, and/or for the spectral analysis as spectral sources.

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