HIGH FREQUENCY CAPACITOR PLASMATORCHES FOR SYNTHESIS OF NEW CERAMIC POWDERS AT HIGH PRESSURE.

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Abstract
The aim of this work is to develop high frequency capacitive plasmatron at high pressure (1 atm) in order to increase the non equilibrium effects produced in that kind of reactor. After a rapid description of the electrical concept of the electrical source and the electrical device needed for this plasmatron we have measured the technical parameters such as electrical field intensity and current for an argon or helium or air plasma working in these conditions. To conclude, electronic temperature, electron concentration, neutral species temperature (enthalpy probe) give us the main properties of this plasma for production of fine powders (0,5 µm) of pure silica).

I - Introduction

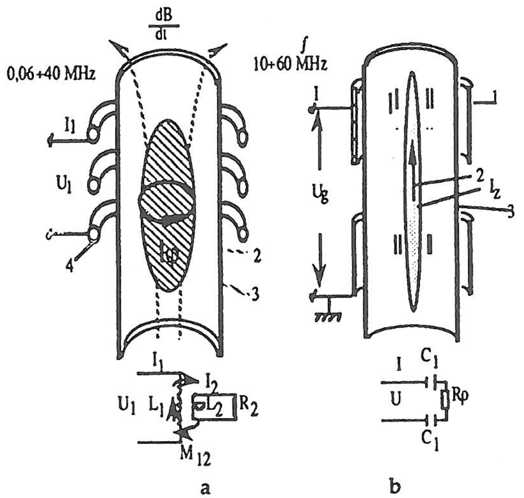


Figure 1 : High frequency inductive plasmatron (a), high frequency capacitive plasmatron (b)

The high frequency induction plasmatorch remained for a long time a single source of the electrodeless pure plasma. This lasted until early 70's when one more principle of generating the electrodeless pure plasma was conceived (1). Now it is embodied in a high frequency capacitive (HFC) plasmatorch. Its operation principle is based on the so-called capacitive coupling of the supply source with the conductive discharge zone. The energy transfer to, the discharge zone is effected by the capacitive current. The outer electrodes are beyond the discharge zone and without contact with the plasma gas. This enables, as in the case of the HF induction plasmatorch, to generate "pure" plasma. Two principles of electrical energy transfer for both HFC and HFI plasmatorches are presented in fig 1.

The electrical and energy coupling of the supply source with the plasma of the HFI plasmatorch is effected by the inductor (4) through varying magnetic field which induces the electrodeless discharge (2) in a conductive gaseous medium (fig 1, a). The supply source in the HFC plasmatorch is coupled to the plasma by the electrical capacitance of the coaxial system formed by two outer electrodes (1), and the plasma string (2). The latter has no direct contact with either the electrodes or the discharge chamber walls, and so the plasma purity is ensured. The HFC plasmatorch plasma differs largely in its nature from that of the HFI plasmatorch. The annular induction current in the HFI plasmatorch is of order of hundred amperes, the plasma is, as a rule, in a condition of thermal equilibrium, and the jet diameter is defined by the plasmatorch diameter and amounts to 5-15 cm. The HFC plasmatorch current is several amperes (1-10 A), the total voltage drop per unit string length is up to 20- 200V/cm, the string diameter is up to 1 cm. This allows to release the large power in the HFC plasmatorch at very low currents (to 10 A). The plasma under such conditions in non-equilibrated even in molecular gases ($T_e > T_{ai}$)

II - The types of HF Capacitive plasmatron

The first attempt to use this type of discharge as a base for the HF plasmatorch was performed in 1960-1961 by one of the authors [2].

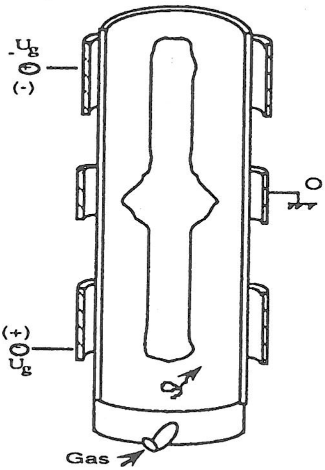
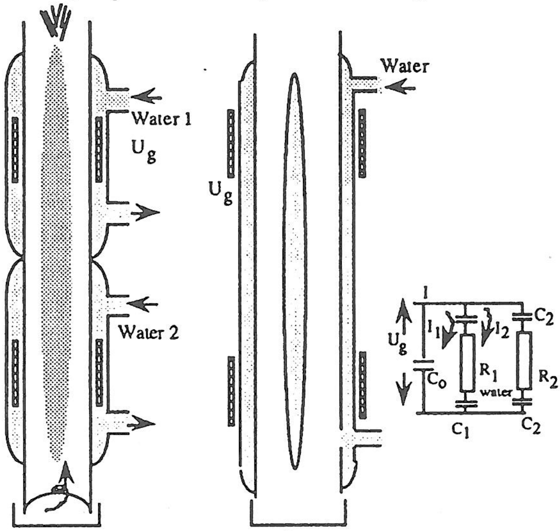


Figure 2 : High frequency capacity plasmatron with three electrodes

In these first articles the axial vortex stabilization of the torch discharge was used for the first time. In order to increase the capacitive coupling of the torch with the earth a grounded coaxial electrode was used.This constituted, in fact, one half of the modern plasmatorch. The plasmatorch of that type are logically termed as linear ones (suggested by the form of the discharge channel extended into a straight line with the coaxial electrodes). The various design modifications of linear HFC plasmatorches are presented in Fig 2. They may employ two or three electrodes (Fig 2). The cylindrical electrodes are placed coaxially in discharge chambers with some gap. The electrode connection to the supply source is constructed so that the potential electrode is situated between the earthed ones. This provides the electromagnetic fields shielding and brings down the jet potential extruding from the discharge chamber. In the case of a multielectrode plasmatorch design several HF-arcs are formed in the discharge chamber correspondingly to the number of electrodes. The discharge current flows along the discharge chamber axis.

The discharge power and current are defined by the capacitive coupling between the electrodes and discharge, which in its turn, depends on the diameter and height. Various designs of the water-cooled discharge chamber (WDC) are shown in fig 3a, b, c,. In fig 3b model the copper annular electrodes are situated outside the WDC. In the design of fig. 3a, the electrodes are placed inside the WDC. That results in more close coupling between the discharge and the electrodes. But this construction is more complicated and calls for the special welding of quartz pipes and current leads. In such designs the current arises between the electrodes through the cooling water. The design of fig 3a, completely eliminates this interelectrode shunt current which is accomplished by dividing of WDC into two water-cooled parts. The optimum design of the HFC-plasmatorch has the electrodes inside the chamber placed closely to the discharge. The chamber is divided into two sections for eliminating the shunt current.The design of fig. 3b, is a basic one for the HFC plasmatorch of linear arrangement. The simplified equivalent electric circuit for the HFC plasmatorch is represented only by the nature of the electric coupling of the discharge with the electrodes. In real designs the distributed capacity C_0 is always present, that is caused by the own capacitance of the electrodes and that of current leads. The equivalent electric is shown in fig.3 c. The HFC discharge being non-uniform in its volume has a laminar structure. One can discern, in particular, the positive discharge column and the electrode layers.



a b c

Figure 3 : High frequency capacitive plasmatron with water-cooled discharge chamber

The energy generation in the layers may affect considerably the plasmatorch efficiency. Following to [3] we shall distinguished the strong-current HFC discharge when the electrode layers are similar to the cathod domain of a low direct-current discharge and the low-current one when the influence of the secondary emission has little effect on the characteristics of the layers, and the discharge current close to the electrodes by the displacement currents. At the low-current discharge the electrode layers are not broken down and the discharge to electrode coupling is purely capacitive. HFC plasmatorches at atmospheric pressure are usually operated under strong-current conditions when the electrode layers are broken down.

III - Experimental measurements

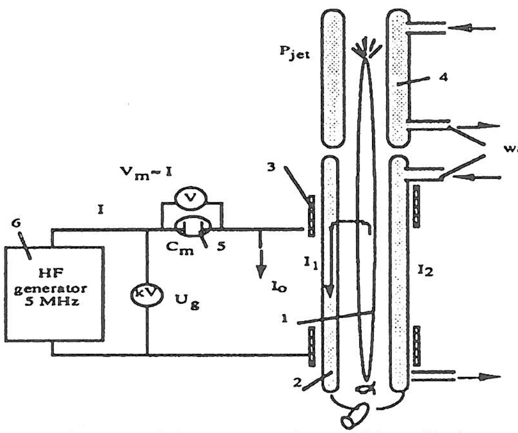


Figure 4 : Scheme experimental installation

The studies of HFC plasmatorches has proved their reliable operation in air, oxygen, helium and hydrogen atmosphere within 13-30 MHz frequency range. One of the ways of increasing the operation reliability of HFC plasmatorches is the decrease of the oscillator frequency. We have obtained a stable discharge at atmospheric pressure and studied the parameters of the HFC plasmatorch at 5.28 MHz.
The local electron and gas temperatures, mean-mass plasma temperature, and electric discharge characteristics were measured. (Fig 4).

HFC discharge (1) was initiated inside the water-cooled quartz discharge chamber (2) of inner diameter 10 mm with the aid of copper coaxial electrodes (3) mounted directely to the outer surface of the discharge chamber (the electrode height was 55 mm, and the diameter (33 mm). The total length of the active plasma string was 210mm. The plasmaforming gas (argon, air, helum) at atmospheric pressure was axially supplied into the discharge chamber.
Both the plasmatorch power P_{jet} and the power in the discharge chamber P_2 were determined separately by calorimeter 4. The controlable voltage was supplied to the electrodes from the oscillating circuit of the valve oscillator 6 (U_{col} = 1 - 7 kV). The current supplied by the oscillator was measured with the aid of the measuring capacitor 5 (C = 225 µF); the current was defined by the voltage drop across the capacitor. The operating frequency was 5.28 MHz (up to 6 MHz). Initiation and maintenance of the HFC discharge at such a low frequency (the earlier studies were performed at f ≥ 17 MHz) were owed mainly to the use of the water-cooled discharge chamber. But some complications of measurements appear, <due to the arising of the shunt current through the layer of the cooling water I_1 There are two currents shunting the main discharge current I in the measuring circuit (fig. 4). These are the current I_0 through the distributed capacitance of the wiring, and the current I_1 through the cooling water layer of the chamber (2). The current Io was defined from the voltage drop across the capacitor C_{meg} at the hollow discharge chamber (without water and discharge), and the current I was defined from the thermal losses in the discharge chamber with water without discharge. In the second case (for zero current I_1) the voltage drop across the measuring capacitance gives the vector sum of current $I_0 + I_1$. But the small phase-shift angle between these vectors (j < 10%) allows to use the algebric sum.The values of I_0 and I_1 are shown in Table 1 for various values of U_{col}.

IV - Results

HFC-discharge in argon is a very complex phenomenon and its spectral diagnostics has not been thoroughly investigated. The spectral diagnostics and some estimative calculations pertaining to the case, are presented in [4]. HFC-discharge is a non-equilibrium phenomenon i.e. the electron temperature is substantially higher than that of the heavy species.

$$I_{cont} \approx N_e^2 T^{-1/2} \qquad (1)$$

For small currents (I < 0.5 A) when the braking continuum of electrons on neutrals is of the same order as a recombination one, the more accurate diagnostics requires the detailed investigation of the role of the continua in the wide range of the wave lengths. In this work such study was not performed.
The electron concentration, defined from the recombination continuum and formula (1) was found to be $N_e \approx 8\text{-}10^{14}$ cm^{-3} (for the current of 1 A). If the braking continuum is relevant (the current I <0.5 A), the electron concentration may be assessed at $N_e \sim 6\text{-}10^{14}$ cm^{-3} according to [5]. The concentration defined from the widening of the hydrogen lines (with the small admixture of hydrogen) $H\beta$ $H\gamma$ by the instrument with small dispertion for the currents I ≈ 1A does not exceed $N_e \leq 1.5.10^{13}$ cm^{-3}.
The determination of the electron temperature of HFC-discharge with reduced errors in this work was effected by comparison of HFC discharge with the high frequency induction discharge having plasma close to the equilibriun state (T_e = 9500° K, N_e = 10^{16} cm^{-3}).

The comparison inspected the ratio of intensities of violet and green lines, as well as the continuum intensity. The first procedure gives the value T = 14000 ± 3000 K, the second one leads to T_e = 15000 ± 3000 K which conforms well with the result of the first method. The relative intensity of close lines does not allow to define T_e with adequate accuracy. The spectral measurements of plasma heavy component temperatures were not performed. The gas temperature was measured by the thermoanemometer (the thermal probe) procedure based on the insertion of a thin (of 0.5 mm diameter) tungsten cylinder into the discharge transversely to the flow. The measurements of local gas temperature by such method are hampered by the fact that the fairly thin discharge string tends to round the thermal probe, and at the large discharge length begins to burn on the probe just as on the electrode. But in a limited range these measurements were still performed. The gas temperature values in argon were found to be $I_r \approx 1200 \pm 150$ K at the discharge axis and 950 ± 100 K at the 3 - 4 mm distance from the string axis. Calculations from the energy balance equation [5] in a very rough estimate give the value Tr = 2000K. The spectral and thermal probe measurements at the string axis of HFC discharge in argon show that the string diameter d depends on the current I has the form $D \approx I^{0.75}$ for d = 1.6 - 2.4 mm, and total radiation intensity J for the different current values is defined by the function $J = I^{1.25}$ (the data are obtained for the spectral photograph section from 410 to 590 nm).

HFC-discharge in a quartz water-cooled chamber considered as an electric load is made up of a complicated system of distributed capacitancies and resistancies. The equivalent circuit presented in fig. 4 reasonably describes the HFC discharge under the conditions of the present paper. Here C_0 and I_0 are the capacitance and current of wiring, I_1 and R_1 are the current and resistance of water, I_2 and R_2 are the current and resistance of the HFC discharge ; C_1 and C_2 are the total capacitancies of two capacitors of the upper and lower electrodes connected in series.

The electrical parameters, mean-mass plasma temperature in argon, air and helium are shown in Tables 2-4, where E is electrical field intensity, P_2 is the power in discharge chamber, P_{jet} is the calorimeter power, T_r was derived from calorimetric measurement ($P_2 + P_{jet}$) by plasma enthalpy. The dependence of currents I_0 and I_1 on the voltage for the investigated plasmatorch is linear. HFC-plasmatorches are of interest to technologies performed in a high-temperature medium (acid, alkaline or neutral) for some peculiarities: lack of contact with electrodes, no replacable plasmatorch pieces (its service life is defined by that .of the oscillator), high field intensity (100 - 200 V/cm), thermal non-equilibrium state of plasma at atmospheric pressure minimum radiation losses, little power required for discharge keeping up and relatively high efficiency amounting to 45 - 60 %.

U_{col}, V	$I_0 + I_1$, A	I_0, A	I_1, A	P_1, W	Notes
2000	0.2	0.09	0.11	57	$R_1 = 6000\ \Omega$
3000	0.29	0.14	0.15	130	
4000	0.38	0.18	0.20	216	$C_1 = 1.43\ \mu F$
5000	0.49	0.23	0.26	355	
6000	0.57	0.28	0.29	532	$C_0 = 1.24\ \mu F$

Table 1

G, g/s	I_{col}, V	I_2, A	P_2, W	P_{jet}, W	R_2, Ω	E_{pl}, V/cm	T_{ai}, K
	6000	1.67	5296	2129	1886	150	1900
1.0	5500	1.28	4335	1860	2624	161	1690
	4950	0.89	3188	1580	4001	170	1480
	6200	1.50	5484	3112	2417	173	1430
2.0	5600	0.79	3423	2451	5515	207	1150
	5200	0.43	2088	1734	11174	230	820

Table 2 : Parameters of HFC plasmatorch in air atmosphere

G,g/s	u_{col} , V	I_2, A	P_2, W	P_{jet}, W	R_2, Ω	E_{pl}, V/cm	T_{ai}, K
0.41		0.6622	238	172	935	29/5	800
0.93	2000	0.7009	239	275	1049	35	570
1.43		0.6677	213	371	1312	42	500
0.41		1.56	517	268	323	24	1250
0.93	4000	1.56	486	394	366	27	800
1.43		1.56	467	472	385	29	630

Table 3 : Parameters of HFC plasmatorch in argon atmosphere

G,g/s	u_{col} , V	I_2, A	P_2, W	P_{jet}, W	R_2, Ω	E_{pl}, V/cm	T_{ai}, K
0.129		0.865	1363	489	2473	102	770
0.295	3000	0.822	777	1175	2743	107	805
0.456		0.725	609	1180	3399	117	525
0.129		1.64	2368	590	1099	86	930
0.295	4000	1.617	1529	1638	1212	93	1110
0.456		1.64	1096	2302	1264	99	930

Table 4 : Parameters of HFC plasmatorch in helium atmosphere

V - Technological applications and conclusion

HFC plasmatron may be applied for plasma chemistry synthesis of ultra fine powders which seem to day a large field for new catalyst, new composit material (polymer an oxide...).
In Russia this installation was made for ultrafine powder of SiO_2 .

(1) High frequency capacitive plasmatron power 10 -15 kW and 13 MHz of frequency
(2) Plasma pressure 1 atm.
(3) Vortex gas flow injection : oxygen
(4) Powder production 1kg/h for particle sizes between 0.2 - 0.5 µm
(5) Gas flow of the starting gas $SiCl_4$ (1.5 kg/h).

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