INSTITUTE OF PLASMA PHYSICS
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# D. C. – ARC PLASMA GENERATOR FOR NONEQUILIBRIUM PLASMACHEMICAL PROCESSES

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ABSTRACT

The analysis of conditions for generation of nonequilibrium plasma to plasmachemical processes is made and the design of d.c.-arc plasma generator on the base of integral criterion is suggested. The measurement of potentials on the plasma column of that generator is presented.

# I. INTRODUCTION

The vibrational energy of molecules $E_v$ (the molecules in the ground or electronically excited states), is the most effective energy in realizing of endothermic high threshold reactions like the simplest one:

$$A+BC \rightleftarrows AB+C \quad . \tag{1}$$

or for the bimolecular diatomic or polyatomic reactions. But the translational energy $E_t$ of the atom and molecule (and mainly of the two molecules) must play a role during the collision and then with the following exchange of atoms (1) etc. From the point of view of the elementary processes it is possible to characterize the reaction by the crossection $\sigma(E_t,E_v)$ depending on both these energies (exactly $\sigma(E_t,E_{vi})$ for $i=1,\ldots,n$ vibrational degrees of freedom of the polyatomic radicals), where the threshold of reaction is given by a threshold curve $E_t=F(E_v)$ situated above the line of threshold energies $E_t+E_v=E_a$ ($E_a$ is the activation energy of reaction ).

The rate coefficient of reaction is obtained by integration of crossection function (from a threshold energies curve) for a given energy distribution functions (eg.) $f_t(E_t),f_v(E_v)$ in the translational and vibrational degrees of freedom respectivelly [1]:

$$K(T_g,T_v) \sim \iint_{E_t=F(E_v)}^{\infty} \sigma(E_t,E_v)E_t^{1/2} f_t(E_t)f_v(E_v)dE_t dE_v \tag{2}$$

This rate coefficient is determined mainly by that threshold curve point where the argument of integral is maximal, i.e. where the sharp maximum occurs for the exponential distribution functions $f_t(E_t),f_v(E_v)$.

In the equilibrium system the equality $T_g=T_v$ is satisfied and the point lays on the threshold curve where the minimum of

energy $F(E_v)+E_v$ occours,i.e in that point where the threshold curve touches the line $E_t+E_v=E_a$.
In the nonequilibrium system the condition $T_g \neq T_v$ occurs and this extreme is found by expression [1]:

$$dF(E_v)/dE_v\ [1-T_g/F(E_v)]=-T_g/T_v \qquad (3)$$

The effective threshold of reaction is not stable in the nonequilibrium system but depends both on the value of translational temperature $T_g$ and on the degree of nonequilibrium $T_v/T_g$ as well.In the case of unlimited growth of $E_v$ (and the croo of $E_t$ ),i.e. in the place where the threshold curve $E_t=F(E_v)$ approaches the $E_v$ axis, the threshold of reaction will be determined as by $T_g$ (the limit aproche $T_g/F(E_v) \to 1$ determines some minimal necessary energy characterized by the translational temperature $T_g$ for the synthesis),as by degree of nonequilibrium $T_v/T_g$.
In the case of lower nonequilibrium $(T_g/F(E_v) \ll 1$. i.e.the threshold curve point is characterized by the higher translational energies and the efective threshold of reaction is determined only by the degree of nonequilibrium $T_v/T_g$.
From these conclusions one can see the importance of reaching maximal degree of nonequilibrium $T_v/T_g$ at the minimal gas temperature $T_g$ for endothermic synthesis.
If the Treanor's distribution holds for the vibrational degree of freedom of diatomic molecules (rather than Boltzman's one)

$$f_v(E_v) \approx \exp\ [-E_v/T_v + x_e E_v^2/T_g\ \hbar\omega\ ] \quad .$$

(where $x_e, \hbar\omega$ are the unharmonicity factor and the vibrational quantum respectively),the shift of the reaction effective threshold is then established (by Matcheret et all [1]),as:

$$dF(E_v)/dE_v\ [1-T_g/F(E_v)]=-T_g/T_v + 2x_e E_v/\hbar\omega \quad . \qquad (4)$$

The exchange and the distribution of energy in the system

under the nonequilibrium conditions occurs at the following ways.

Under the laser pumping of a diatomic molecules ($h\nu$-V processes) the pumping affects in general on the first few vibrational levels only and the absorbed quanta are then redistributed by the vibrational up-pumping Treanor's mechanism [2] (V-V proc.).The dissociation can occur when the introduced quanta reach a level located above the last bound vibrational level of the molecule (V-D).

Under the electrical discharge the excitation of vibrational levels is operated by the free electrons (e-V) from a ground level (or from a higher ones),depending on the energy of free electrons.The free electrons can dissociate (e-D) the molecules independently from the vibrational up-pumping mechanism (V-V).On the other hand the creating atoms can effectively destroy the vibrational content of the molecules.then the low residence time of molecules in the discharge (for the continuous discharges) is supested [2]. The loss of vibrational quanta by dissociation (V-D) and the spontaneous emission (V-$h\nu$) can occurs in the system too. The most important processes one can expect in the redistribution of vibrational energy to the translational one (V-T). The gas temperature $T_g$ strongly affects the vibrational distribution. The increas of $T_g$ increases V-T exchange (and decreases V-V ones).then progresively shifts the Treanor's distribution to the Boltzman's one and eliminates the nonequilibrium effects (moreover in the case of unharmonic oscilator V-V exchange tends to heat the gas too).For the gas temperature $T_g \cong 5kK$ can V-T processes overcome V-V up-pumping mechanism and $h\nu$-V ones [4] From these conclusions one can see the importance of keeping the gas temperature $T_g$ as low as possible to emphasize the nonequilibrium effects. For the reactions of polyatomic molecules in afterglow the energy of depopulation of vibrational distribution is moreover significantly lower than for diatomic ones [3].

Under the electric discharge the dissociation constant $K_D$ of diatomic molecules through the pure vibrational mechanism

(PVM) and the direct electron (impact) mechanism (DEM) is possible defined as a function of reduced electric field E/N. In the glow discharge of nitrogen $N_2$ the experimental results of measurements of $K_d^{max}$ constants are in good agreement with the theoretical ones in the range of values $E/N > 6.10^{-16}$ Vcm (for DEM), while this value is an upper limit for PVM.
In the glow discharge of hydrogen $H_2$, the atoms formed by DEM from $b^3\Sigma u$ state can sufficiently destroy the vibrational content (at $T_g > 300$ K, $n_e < 10^{11}$ $cm^{-3}$ and $E/N > 3.10^{-16}$ $Vcm^2$) and the vibrational quanta introduced by e-V processes cannot reach the top of the vibrational ladder (the H atoms in concentration $[H] \geq 10^{-3}.[H_2]$ strongly reduces the importance of PVM) [2].

If an average energy of free electrons in discharge is low ($0.5 < \bar{\varepsilon} < 3$ eV), then the loss of their energy through the vibrational inelastic collisions preferentially occurs [4]. These collisions populate the vibrational levels of molecules which are then able return the part of energy back to electrons by second-kind collisions (V-e). Consequently, the electron energy $\bar{\varepsilon}$ can increase by a value corresponding to the vibrational energy $\bar{\varepsilon}_v$ and this energy can then represent a source of power for the free electrons. The coupling between the electron edf and the vibrational distribution $N_v$ of the ground state of the molecules then occurs. During relaxation in the afterglow the elastic vibrational losses and the superelastic vibrational gains balances themselves in the time $\tau \approx 10^{-7}$ s, then a quasi-stationary conditions with the Treanor's $N_v$ distributon (or similar one) creates and with Maxwell's electron edf. The electron temperature $T_e$ approches the vibrational $T_v$ (both beeing higher than the gas temperature $T_g$). This state can be changed by the variation of vibrational distribution in the time of order $(10^{-3}-10^{-2})$ s as a result of V-V and V-T processes [4].
From the previous follows that the nitrogen discharge (without traces of hydrogen) is, if possible, demanded.

To compare the theoretical and experimental results, Capitelli et all [4] established the criterion under the hypothe-

sis that when a quasi-stationary state between $T_v$ and $T_e$ is in the post-discharge achieved.the characterization of conditions must pass through the characterization of the initial $T_v$ values.The temperature $T_v$ is then a function of E/N. $n_e$. $\tau$ and p ($\tau$ is the residence time in the discharge.p is the pressure) and at the initial (nonstationary) conditions $T_v$ increases with increase of $E/N.n_e$,$\tau$ and decreases with p.Then some q factor characterizes the value of $T_v$ (as a linear dependence) in the discharge is $q=(E/N)\ n_e\ \tau/p$ [4].
For a low pressure discharge of $N_2$ (p < 5 Torr) the initial theoretical and experimental conditions characterized by the similar q factors ($q \approx 10^{-7}:10^{-6}$ for $T_v$=8.5; 3.5 kK respectively).were in good agreement with the temporal evolution of energy $\bar{\varepsilon}(t)$ and then with $T_v$.
For a low pressure discharge of $H_2$ the temperature $T_v > T_e$ (the superelastic gains must compensate the inelastic vibrational losses with elastic and rotational ones being not negligible) [4].

The coupling between the vibrational distribution function of the ground state ($N_v$) and that of electronically excited states ($N_v^*$) concerns the high-lying vibrational levels occurs in the discharge too. For a long times (t > 1 ms) of the nitrogen afterglow the created high-energy electrons (V-e) can completely alter the situation known in range of microseconds and strong coupling between $N_v^*$ and eof can be another complication for solving of the problem [4].

If an nonequilibrium given by the values $T_v/T_g$ and $T_g$ is in the system the relaxation of system together with the endothermic reaction in afterglow can be plotted by the motion of a point on trajectory in the phase plane (or space), determined by coordinates $T_v$,$T_g$ [1].Because of the essentially different relaxation and reaction velocities an areas when the relaxation or reaction prevales is possible determine in this phase plane (the other process is negligible in area because of the exponential character of functions). The relaxation boundary for the system without reverse reactions is determined between areas.

In the case the energy introduced to the system determine the initial conditions above the relaxation boundary the reaction prevales and the motion of the point to this boundary will largely given by the losses of $E_v$ (provided the loses from the diferent vibrational levels already don't reach up to the enthalpy of reaction).As far as the relaxation boundary is reached the relaxation starts compete with reaction at the constant energy content of the systeme ($-\Delta E_v=\Delta E_t$) and the trajectory is the line of constant energy. If the relaxation is faster (on the dynamics of the systeme depends), the competition is to the smoller content of energy of the system (to the lower temperature $T_0$) as far as the ecuilibrium $T_v=T_0$ is reached [1].

The reaction efficiency is conected both with the excess of energy $\sim(T_v+T_0)$ over the reaction enthalpy introduced to the system as on the excess of the vibrational energy $\sim(T_v/T_0)$ during V-T relaxation.The nonequilibrium reaction is finished on some ecuilibrium temperature $T_{ec}$ given by the energy threshold of reaction and the dynamics of the systeme. If $E_\infty$ designates the introduced energy the integral reaction efficiency is then given by [1]:

$$\eta = 1-T_{e0}(c_v+c_t)/E_\infty \quad , \tag{5}$$

where $c_v$,$c_t$ are the vibrational and translational specific heats respectively.

The conclusions follows from the previous that the increas of efficiency is posible due to the increasing of introduced energy $E_\infty$,the reaching as low ecuilibrium temperature as possible (the reaching as high nonecuilibrium $T_v/T_0$ as possible). In the choosing of reactants with smolest specific heats (the gase phase reactants choose only),and due to the insulating of systeme with respect to get slower V-T relaxation or deactivation on the reactor wall.

The thermodymamic reason is for synthesis in nonecuilibrium system too because the synthesis (and condensation) occurs at the gas temperature $T_0$ much lower than the tempera-

ture of dissociation and excitation of reacents under the reaction [5,6].

From the discharge theory it is known [7], that the energy transfer from electric field is essentially due to the free electrons and increase of the electric field intensity E results in growth of their energy. The electrons excite (ionize,dissociate ...) heavy particles under the collisions.
In the case of conservation of electron gas energy (in the weak electric field E) the estimation of degree of temperature nonequilibrium is possible by the Finkelburg criterion (the exchange of energy is due to the elastic collisions only and the electron drift speed is proportional to E), comparing the mean drift energy of electrons with their temperature energy [8,9].

$$(T_e - T)/T_e = (M/4m_e)\,(\lambda_e eE/(3/2)kT_e)^2 \quad . \qquad (6)$$

where $\lambda_e$ is the mean free path of electrons, $m_e$ is the mass of electron.
In the strong electric field will be estimation more complex because of the contribution of inelastic collision, but for qualitative estimation the criterion is still universal.

The number of particle collisions at reaction have to be maximal in order to maximizing the efficiency of reaction. If the growth of particle concentration (then growth of pressure p ) is required,then mean free path $\lambda_e$ will be shorter. If $\lambda_e$ is constant (e.g.at atmospheric pressure), the nonequilibrium is possible get higher by decreasing of the mean energy of electrons (the kinetic energy of thermal motion) or by the increasing of electric field intensity E.
The linear d.c.-arc generator with the transpirational discharge channel wall is proper use to reach extremes of both these quantities,in which the strong influence of cold gas over the all discharge length occurs.At this type of generator the electric field intensity E increases approximately exponentially with respect to the length of the discharge channel and to the gas flow rate. The field intensity can

reach the value of E=250 V/cm at the air discharge (in contrast to the value of E=60 V/cm only in the smooth wall channel generator of similar type)[10]. The dependence for nitrogen $N_2$ is similar [11].

The breaking of the LTR in the argon arc plasma was theoretically studied by the two temperature model of smooth wall channel (the parabolical profil of velocity v ) for the currents I=(5÷160) A, flow rates g=(0.5÷5) g/s, channels radii R=(0.5;1) cm, and at pressures p=(0.01;0.1) MPa [12]. While the nonequilibrium was found in all cases of the model at the lower pressure the nonequilibrium on the axis of the arc was found at the currents lower than I=10 A at the atmospheric pressure. The nonequilibrium $\Delta T \simeq 1$ kK ≃ const on all the radii r (except that near the wall, where increases sharply) in the origin of the channel at the current I=5 A occurs, while at the length z/2R=10 of the channel the nonequilibrium increases from the radius r/R≃0.3 already. The increase of flow rate in this type of channel influence on the elongation of nonequilibrium zone (with growth of the arc temperature due to the growth of pressure). The increase of channel radius in the model tends to the equalization of temperatures because the constant flow rate results to smaller specific flow rate (and by the absorption of the radiation perhaps).

The model results are compared with the another experimental ones where states the nonequilibrium can appears at currents I≃50A or at parameters I/2R < 100 A/cm. The nonequilibrium can reach value $\Delta T$ < 1 kK on the axis of the argon arc [12].

From previous follows the lower currents I, the smaller channel radii R, and higher flow rates g, are demanded to make higher the degree of nonequilibrium.

The parameter E/p is possible use as parameter of nonequilibrium because the mean drift velocity of particles between collisions is proportional to this parameter, as was stated. Prender states the boundary between the thermal and nonequilibrium plasmas as E/p > 10 V/mkPa, while the quantity can exceed value $10^4$ V/mkPa in nonequilibrium ones [13].

## II. CRITERION OF NONEQUILIBRIUM

From the volt-ampere characteristic of the arc discharge one can see that the same value of E (at p=const) can discharge reache for two or more currents I. The values of conductivity $\sigma$ of the plasma differs in these cases. The nonequilibrium is possible increase by decreasing of the thermal energy of electrons then by decreasing of the current I.

The nonequilibrium one could appreciate appearently by the comparison of the characteristic times $\tau$ of excitations of individual degrees of freedom with the characteristic times of relaxations and reactions only.Capitelli's criterion (expanded Pfender's) takes more into account this reality by the quantities of electron concentration and residence time of the molecules in the discharge.The integral quantities and that concerning of the elementar processes are in the criterion as well.

On the base of preceding results the nonequilibrium in discharge characterized by the quotient $T_v/T_g$ ( or $T_e/T_g$ ),with the quantity of gas temperature $T_g$ (rather than any difference $\Delta T$),is possible determine by the easy measurable integral quantities E,I,p,v,only,as:

$$\bar{q} = (E/I)(v/p) \quad [\ \Omega/s.Pa\ ] \ . \tag{7}$$

for the flowing plasmas. From the criterion follows that the conductivity of plasma I/E is necessary decrease thereby decrease the mobility of electrons $\mu_e$ and the current density $j_e=n_e e\mu_e$. To reach the higher nonequilibrium ($\bar{q} \sim T_e/T_g$), the residence time in the discharge is necessary get shorter from the macroscopic point of view.

From preliminar calculations it is seen that the parameter $\bar{q}$ can reach the values in the range $10^{-4}<\bar{q}<10^{3}$, but dependence on the degree of nonequilibrium,as well as on the boundary of nonequilibrium ($\bar{q}\approx(10^{-3}+10^{-2})$) will be necessary studied in the future.

This criterion is valid for the sufficient number of colli-

sions of particles with the electrons in the discharge.In the opposite case the characteristic times $\tau$ are necessary consider,as was said.

The nonequilibrium can be increased by decreasing of the current I. The forward rate coefficients of reactions then can increas and reverse decreas with respect to the rate coefficients of reactions at equilibrium synthesis. [1,2]. The best conditions for syntheses are at the lowest power of the given device,what is a well known fact in UHF syntheses [14]. Because of the electric field intensity E has to be as high as possible in the case of nonequilibrium plasma (and current I minimal),then it is not convenient characterize the generated plasma by the power or the enthalpy. The physical parameters of that plasmas can differ substantially [15] from the point of view of synthesis.

## III. EXPERIMENTAL,RESULTS

The maximal values of E,v,is necessary seek for at the design of the d.c.-arc plasma generator for nonequilibrium plasmachemical processes (syntheses) at atmospheric pressure (see(7)).The flow rates q are not as important as velocities v because of decreasing the thermal energy of electrons and particles by the mixing rates. The current I has to be minimal.From the previous follows that the transpiration channel wall generator is necessary and the radial (and/or tangential) components of velocities must stabilize the minimal current of the discharge.

The particle concentration $N \sim 10^{18} cm^{-3}$ is in the nitrogen atmospheric pressure discharge,then for $E \geq 250$ V/cm the reduced electric field $E/N \geq 10^{-16}$ V/cm is possible reached. The FVM will still prevale in the discharge and one can expect the electron concentration $n_e \leq 10^{13} cm^{-3}$ at the temperatures of the gas $T_g \geq 6$ kK.

The arc-generator of this type has been designed. The discharge channel radius of generator is R=0.4 cm and the length of the column is approximately $l \geq 6.5$ cm with dependence on

the length of anode (Fig.1.)

The start of arc between the cathode and assistant anode occurs by the high-frequency,high-voltage circuits. The arc then switches over on the anode (16).The potentials along the channel are picked-up by the ring probes which introduce the gas flow into channel simultaneously. The working medium has been argon Ar up to date.The measured potentials for the flow rate g=2.2 g/s of the argon are in Tab.1.
The values are in agreement with the values measured at the pore-like transpirational wall generator (1).

## IV. CONCLUSIONS

The activation energy $E_a$ of endothermic synthesis can be very high.To estimate this energy by Polanyi-Semenov's rule it is possible as $E_a$=48.12+0.75 Q in equilibrium case (17) (where Q is the enthalpy of reaction), or by the dissociation energies of reagents only. The enthalpies Q can reach the extreme value for synthesis of nitrides (Q=7.5 $10^2$ kJ/mole for $Si_3N_4$ (18)). The primar interest is to get lower the thresholds of these reactions, hence to reach the high nonequilibrium $T_v/T_g$ at low gas temperature $T_g$ in the d.c.-arc. In the nitrogen discharge can still PVM prevale for the value of E/N≅2.5 $10^{-16}$ $Vcm^2$,and temperatures ($T_v \simeq T_e$) > $T_g$ are supposed in the afterglow. The efficiency of nonequilibrium reaction can then be higher.
The criterion of nonequilibrium is established by integral quantities and on the base of it the design of d.c.-arc generator is proposed. The measurements of some potentials on the plasma column are given. The values of E are a good promise for the future work.

The endothermic reactions in the arc discharge, or in the afterglow of this discharge, has been studied at the currents I > 100 A up to day, although it is well known that the increase of currents decrease the nonequilibrium of the system (10,19,20).The higher effectivity of these reactions requires next decreasing of the discharge current with simultaneous

increasing of the degree of nonequilibrium, although the generators will operate in the region of negative differential conductivity.

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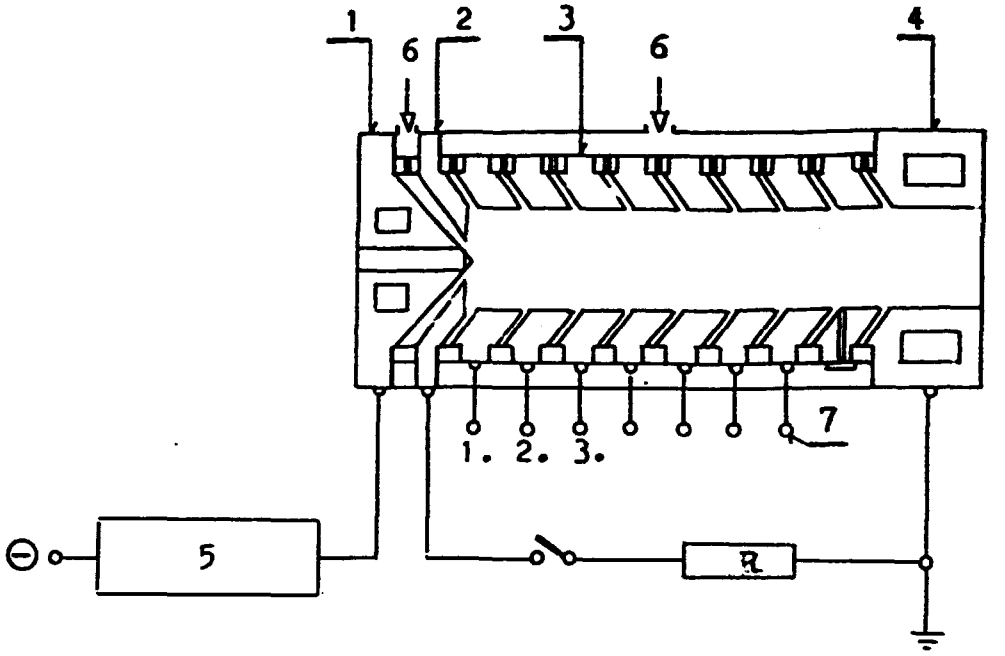


Fig.1. The arrangement of d.c.-arc plasma generator. 1 - Cathode; 2 - assistance anode; 3 - ring section (metal.and insul.rings); 4 - anode; 5 - H.F.-H.V. circuits; 6 - gas inlets; 7 - potential terminals. Argon flow: U=90 V; I=48 A; $p \cong 10^{5}$ Pa; $g=2.2$ gs$^{-1}$; $\bar{g}=(10^{-3} \div 10^{-2})$.

**Table 1**

Potentials $\Delta U$ and electric field intensity E on the plasma column

| N° of term. | 1.-3. | 3.-5. | 5.-9. | 9.-13. | Cond. |
|---|---|---|---|---|---|
| $\Delta U$ [V]- | 8.75 | 9.75 | 19.40 | 20.30 | 1 |
| E [Vm$^{-1}$] | 975 | 1080 | 1080 | 1120 | |
| E [Vm$^{-1}$] | ≈600 | | | | 2 |

Conditions -1 : I=48 A; $g=2.2$ gs$^{-1}$; $2R=8.10^{-3}$ m; $L/2R=8$

-2 (11): I=60 A; $g=1.1$ gs$^{-1}$; $2R=10.10^{-3}$ m; $L/2R=5$