Dissociation of molecules in plasma and gas: the energy

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Abstract – Various physical mechanisms of dissociation of
molecules in plasma and gas are observed. Experimental data
on energy cost of atoms for molecular dissociation at diver-
se conditions are analyzed. Maximal attainable values of
this parameter for molecular dissociation in gas discharge,
at electron beam irradiation of gas, in result of photodis-
sociation of molecules, at a thermal heating.

INTRODUCTION

Many chemical processes in a gas or a plasma proceed with a partici-
pation of atoms or radicals. In order to accomplish this process one needs
at the first stage to break the molecule into components. Since there are
many secondary processes in the considered manner of energy insertion to a
system, to form free atoms one needs to introduce into the system a higher
energy than that necessary for a molecule dissociation. An energy characte-
ristic describing efficiency of the considered way for energy introduction
to the system for dissociation of molecules, is an energy cost of atom or
radical $\mathcal{E}$. This value is the ratio of energy introduced into the system
to the total amount of atoms formed. By comparing various ways of molecule
dissociation consumptions of energy one can valuate an efficiency of a cor-
responding method of energy insertion. This analysis was performed in the
present review (see also [1]).

Among various ways of molecule dissociation in gas and plasma we shall
consider an atom generation in a gas discharge, under the effect of an elec-
tron beam and ultraviolet radiation, as well as while heated a gas in an
arc discharge or near a heated surface. These are the main ways of disso-
ciation of molecules. We have some amount of the needed information for a
limited number of molecules, such as fluorine, oxygen, nitrogen, carbon
dioxide, which are concerned with certain applied problems.

Let us note that an energy cost of atom generation exceeds always an
energy of breaking of a corresponding molecule bond, since along withe dis-
sociation of molecules a wide variety of other processes proceeds in the
systems; in particular, an excitation of vibrational and electron states
of molecules. A major portion of energy inserted initially into a gas is
consumed by these processes. As a result of relaxational processes this
energy is transformed to a heat. Thereby the energy cost of atom formation
characterizes also a degree of gas heating at a corresponding efficiency
of the process. This circumstance is the most essential in the event when

the process proceeds at a low temperature; it confines the process potentialities.

Information on an energy cost of atomic fluorine formation from a molecular one in a variety of ways is collected in Table 1. Considerable information on this subject is connected with the use of an atomic fluorine in chemical HF-lasers as well as to perform nonequlibrium chemical processes with formation of fluorine metastable compounds of the KrF_2 type.

Table 1

Energy cost of fluorine atom formation

Way of influence	Energy cost, eV	References	Notes
Gas discharge	6–8	[2]	A glow discharge in a molecular fluorine
	60	[3]	UHF- discharge in a mixture: $Ar:F_2(10:1)$, fluorine dissociation 90%.
	3–40	[4]	Glow discharge, fluorine pressure 0.25–5 Torr, current density up to $60mA/cm^2$, temperature – from room to 680 K.
	3	[5]	UHF- discharge in fluorine, pressure 32 Torr, input power 0.1–2 kW, frequency 8mHz.
Electron beam	4,5	[6]	Electron energy 120KeV, mixture $SF_6:H_2(11:1)$
	6	[7]	Electron energy 120KeV, mixture $He:Ar:F_2:H_2(54:34:6:6)$, cost of a pair of charged particles 24 eV.
	10	[8]	Electron energy 40–50 KeV, mixture $He:F_2:HCl:CO_2(94:4:1:1)$, 3–4 fluorine atoms are formed per every pair of charged particles
	5	[9]	Electron energy – 1 MeV in molecular fluorine
Ultraviolet radiation	1.1– –2.5	–	Maximum cross-section of photodissociation $F_2(2 \cdot 10^{-20}$ cm$)$ conforms with a wave -length 0.3μ which an energy atom cost 2.1 eV corresponds to.
Heated surface	3–5	[10–12]	Temperature of nickel surface about 900 K, fluorine pressure 10–50 Torr.

Referring to Table 1, let us analyze various ways of molecule dissociation. In gas-discharge facilities an energy cost of atom can significantly depend on a sort of gas or type of discharge. This has to do with the fact that the process of molecule dissociation by an electron impact involves a molecule excitation to a repulsive level and subsequent decay of the molecule. Therefore, this process if of a threshold character and its efficiency depends strongly on a mean electron in a discharge. However, even under optimum conditions the energy cost of atom exceeds markedly a half of

molecule dissociation energy.

In gas dissociation by an electron beam a certain portion of electron energy is consumed by dissociation of molecules. Since molecular dissociations takes place as a result of secondary processes after passing a fast electron, the energy cost of atom in this case depends not strongly on the electron energy in the beam and is in several times excess of the molecule dissociation energy. Thus, according to Table 1, the averaged ratio of the energy cost of fluorine atoms formation under the effect of an electron beam to the energy of fluorine molecule dissociation amounts to $3.8^{\pm}1.3$.

A photodissociation of molecules is capable of providing the least values of atom energy cost. This is concerned with the fact that in photodissociations of molecules, secondary processes which consume photons are absent in gas. For that reason the photodissociation of molecules is a convenient way to obtain atoms in a gas. But at high intensities of the process the use of radiation to obtain atoms generate a variety of additional problems which reduce potentialities of this method and make it less convenient in comparison with others.

In Table 1 energy costs of fluorine atom formation in a molecular fluorine are compared at different ways of gas excitation. As it is seen, in the discharge this value depends drastically on the discharge parameters; this value for other ways of energy insertion in gas can be predicted before hand with an accuracy not worse than 50%. Let us note that the energy of fluorine molecule dissociation is equal to 1.63 eV, i.e. the minimum energy needed to form one fluorine atom amounts to 0.82 eV. In all cases an essentially higher energy is consumed for this.

Since the molecule dissociation as result of irradiation of gas by an electron beam proceeds under the action of secondary electrons, which initial energy is in several fold excess of the molecule ionization potential, one can expect that the order of magnitude for the energy cost of fluorine atom will be kept either at the substitution of fluorine molecule by other fluorine-containing molecules, or at changes in a gas composition. Taking into account this circumstance we included in Table 1 a cost of fluorine atom formation from SF_6. The measurements conducted in [13] give the value 6.5 eV for CF_4 and 5.6 eV for C_5F_{12} for the energy cost of fluorine atom formation at the gas excitation by the electron beam, provided the value 4.5 eV is taken as an energy cost of fluorine from SF_6. Naturally, these figures will be varied depending on a mixture composition, but, as it is seen, all obtained figures lie in not a very wide interval. Taking this into account and making statistical averaging of the given results one can recommend the following value for an energy cost of fluorine atom at the dissociation of fluorine-containing molecules by an electron beam:

$$E_F = 6 \pm 2 \text{ eV}$$

This corresponds to the efficiency of molecule dissociation (i.e. the portion of input energy consumed by dissociation) $17^{\pm}4\%$.

As is seen from Table 1, the energy cost of fluorine atom formation in the discharge depends significantly on discharge parameters. The mechanism of fluorine dissociation in the discharge was analyzed by D.J.Slovetskii [14, 15]. Based on the sum total of data he claims that the fluorine dissociation in a discharge can be explained neither by the dissociative

attachment of electrons to a molecule, nor by the step excitation of vibrational levels of the fluorine molecule ground electron state. In these cases the effective constant of dissociation rate should increase with the growth of pressure, what is contrary to observed dependences. The dissociation in the molecular fluorine discharge is a result of an electron excitation of molecule to a repulsive term. This statement agrees with the physics of process, because the fluorine molecule has several low-lying repulsive terms (see, for example, [16]).

Table 2

The energy cost of dissociation of CO_2 ($CO_2 \rightarrow CO + \frac{1}{2}O_2 - 2.90$ eV) molecule in a glow discharge* at $E/p = 24$ V/(cm.Torr) [17]

Relation between amount of CO_2, N_2 and He in mixture	K_{diss}, cm^{-1} $Torr^{-1}$	Energy expended in destruction of one CO_2 molecule, eV
100:0:0	0.35 ± 0.5	68^{+12}_{8}
4 :4:92	6.7 ± 1.5	3.6 ± 0.8
6:12:82	5.5 ± 0.5	4.4 ± 0.4
7:7:86	3.4 ± 1	$7.1^{+2.9}_{-1.7}$

*Energy ξ expended in dissociation of one molecule is expressed through a constant of molecule dissociation rate K_{diss} by an electron impact in discharge, by the relation $\xi = eEw/NK_{diss}$, where E is the electric field strengh in a discharge, w - is the electron drift velocity, N - is the density of molecules.

The sharp dependence of atom energy cost on discharge parameters is also observed in the cases of other gases. Values of this characteristic at various composition of the mixture for CO_2 dissociation are given in Table 2. An addition of He to CO_2 increases the electron mean energy in the discharge, which with a sufficient CO_2 concentration leads to the decrease in energy expended in dissociation of an molecule. In paper [18], where a dissociation of CO_2 in a glow discharge in the mixture CO_2:He (40:10) was a studied, the energy expended in one CO_2 molecule dissociation amounted to 113 eV (under these conditions, 23% of input power is expended in dissociation).

The effective dissociation proceeds under step the excitation of molecules when the breaking of molecule is accomplished from excited states. This is feasible at a sufficient input discharge power and electron density in the discharge, if collisions of one molecule with electrons happen frequently enough. Values of the energy cost of nitrogen atom formation in nitrogen are given in Table 3 for various vibrational temperature. The data are based on calculations [19]*)

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*) Glow discharge investigations carried out in paper [13] for a pure CO_2 have shown that the dissociation involves an electron excitation of CO_2 molecule into one repulsive terms: $e + CO_2 \longrightarrow e + CO_2(A^1\Sigma) - 7eV \longrightarrow e + CO + O$, $\longrightarrow e + CO_2(a^3\Pi) - 10.5eV$ Minimum expenditures in one CO_2 molecule dissociation under optimal conditions for a glow discharge, come up to 10 eV per molecule.

Table 3

The energy cost of nitrogen atom formation at the dissociation of nitrogen molecules in the discharge according to calculations [19] for E/N= $6 \cdot 10^{-16}$ V cm^2

T, K	300	2100	4900
Energy cost of atom, eV	370	220	100

As is seen, with the increase in vibrational temperature the energy cost of atom formation decreases significantly, since in this case the dissociation of molecules is facilitated. But the energy cost of nitrogen atom formation itself exceeds many times the molecule dissociation energy. Such is the case in CO [20,21]. This is primarily explained by a high dissociation energy of N$_2$ and CO molecules, so that it exceeds considerably the electron mean energy and the dissociation process can prove to be multistage. Besides, electron - excited states of N$_2$ and CO molecules, which are excited effectively by an electron impact, are stable and do not result in the molecule dissociation [22]. Thereby, the gas discharge is not suited for dissociation of N$_2$ and CO.

However, for gases with a moderate dissociation energy the gas discharge is the effective means of atom generation. This can be illustrated on the example [23] where oxygen dissociation was performed. Such a process was accomplished on UHF-discharge (2.45 10^9 Hz) with input power 1kw. The oxygen (21%) - helium (79%) mixture was introduced into this discharge at a pressure of 5.2 Torr. The energy cost of oxygen atom formation was 11eV, i.e. the oxygen dissociation consumed 23% of the discharge input energy.

In this case, 18% of the energy was expended in heating the gas, and 50% of the input energy was radiated. As is seen, over a half of the discharge energy absorbed by gas consumed by the oxygen dissociation.

A thermal dissociation of molecules is of specific interest. The energy cost of $\mathcal{E}_A$ atom formed due to the thermal dissociation:

$$AB \rightleftarrows A + B \qquad (1)$$

accounts for:

$$\mathcal{E}_A = D + \frac{[AB]}{[A]}\int_{T_o}^{T} C_{AB}\, dT + \int_{T_o}^{T} C_A\, dT + \frac{[B]}{[A]}\int_{T_o}^{T} C_B\, dT, \qquad (2)$$

where D is the molecule dissociation energy, T_o is the initial temperature; C_x is heat capacity for a corresponding gas component; $[X]$ is the density of the given sort particles. Formula (2) takes into account that in order to decompose molecule we heat-up gas up to a certain temperature, so that the energy is expended in heating atoms and molecules as well as in breaking bonds in a molecule. For two-atomic molecules consisting of the same atoms (A$_2$⇄2A) this formula changes to the shape

$$\mathcal{E}_A = \tfrac{1}{2} D + \frac{[A_2]}{[A]}\int_{T_o}^{T} C_{A_2}\, dT + \int_{T_o}^{T} C_A\, dT. \qquad (3)$$

From this formula it is followed that the using of present method at a sufficient heating provides the cost of atom formation, being less than molecular dissociation energy. Indeed, a noticeable degree of dissociation is attained at low thermal energies of particles as compared with the energy

of dissociation of molecules ($T \ll D$). Since $C_{A_2} \sim C_A \sim 1$, from this is follows that within this temperature range the second and third addenda in formula, (3) or three last addenda in formula (2) make a small contribution to the energy cost of atom. Values of the fluorine atom energy cost are given in Table 4, calculated from formula (3) with allowance for equilibrium concentrations of an atomic and molecular fluorine. As it is seen, at not very low temperatures the energy cost of fluorine atom is comparable with the energy of molecule dissociation.

Table 4

The energy cost of fluorine atom formation, eV, at the interaction with a hot surface ($F_2 \rightleftarrows 2F$ =1.63 eV)

Fluorine pressure, Torr	T,K				
	800	900	1000	1100	1200
10	2.9(175)	1.55(26.6)	1.21(6.76)	1.05(2.72)	1.04(1.69)
100	7.1(24.4)	2.82(6.02)	1.70(2.25)	1.22(1.39)	1.18(1.25)

Along with the thermal dissociation of molecules, secondary processes take place, lowering the energy cost of atom. In particular, when a dissociation of molecules occurs on a heated surface, the energy is expended not only in heating a gas and dissociation of molecules but in radiation of the surface. Values of the energy cost of fluorine atom at the F_2 thermal dissociation near a nickel heated surface are given in Table 4 in parentheses. In this case, the surface is believed to radiate as an black body of a given temperature and an atom formed near the surface is used, not returning to it any more. For probability of dissociation a fluorine molecule colliding with the heated nickel surface, use was made of formula $\boxed{10}$ P=1.9 · ·10^5exp(-17 800/T). Let us note that under realistic conditions the atom energy cost is higher than that of the mentioned one, since together with dissociation of molecules there takes place recombination of atoms on the surface. In doing so, the energy cost of atom formation depends on particular conditious in a chemical reactor.
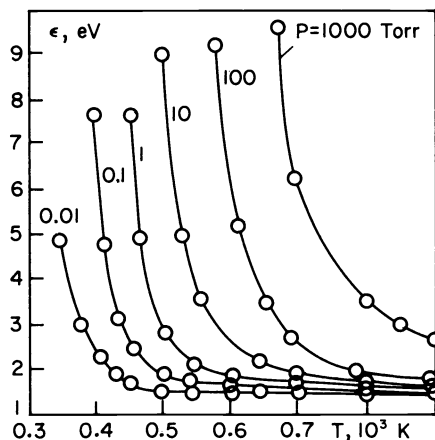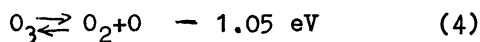

Fig. 1. Dissociation energy as a function of the temperature

Fig. 1 shows another example of the considered type, concerned with dissociation of ozone near a heated surface:

$$O_3 \rightleftarrows O_2 + O \quad - 1.05 \text{ eV} \qquad (4)$$

The surface temperature is lower, so that the heated surface radiation is less significaut. Ozone and atomic oxygen on the heated surface are in a thermodynamic equilibrium.

By analyzing the thermal method of atom formation as a result of dissociation of molecules near a heated surface, it should be noted that it is convenient at not very high temperatures of the surface. High temperatures lead to a strong surface radiation, as well as to its degradation

as a result of material evaporation and chemical processes on the surface. Therefore, the thermal way of molecule dissociation near a heated surface is suitable only for molecules with a moderate energy of dissociation.

Nevertheless, the manner of atom generation itself as a result of a high heating a gas is reather effective. The gas heating is convenient to perform if an electron current is passed through a gas - in a gas discharge, in an arc discharge, etc. In contrast to a case of a heated wall, in this case the heated region radiation can play no part, since a heated gas (or a plasma) in a main frequency region in optically transparent. Atoms generated in a gas hot region diffuse to a cold one where can be used. Tables 5-7 give atom concentrations and energy costs of atom formation, corresponding to heating the simpliest diatomic gases under conditions of a thermodynamic equilibrium in it. The cost of atom formation was determined from formula (3). Here, $C_A = 5/2$ and it was assumed that at temperatures under consideration a molecule is highly vibrationally excited and its heat capacity $C_A = 9/2$, so that formula (3) takes the form:

$$\mathcal{E}_A = \frac{D}{2} + \frac{9}{2} T \frac{[A_2]}{[A]} + \frac{5}{2} T. \qquad (5)$$

Thus, based on Tables 5-7 we can conclude that by heating a gas with the aid of an equilibrium plasma and by keeping in it (gas) a thermal equilibrium, it can be dissociated with a high efficiency. A specific problem lies in atom transport to a place where they are used; and this process will be require an additional energy expenditures. As for generation of atoms, then within the considered pressure range of 0.1-10 Torr for H_2, N_2 and O_2 a practically complete dissociation comes at $T/D = 0.06-0.07$ (T- a temperature, D - an energy of molecule dissociation; in this case, a portion of input energy expended in heating a gas for dissociation of molecules, exceeds 70%).
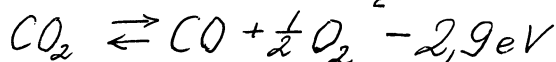
Table 5

The energy cost of formation, eV, and concentrations of hydrogen atoms, % (in parentheses) for heated hydrogen under conditions of thermodynamic equilibrium $H_2 \rightleftarrows 2H$ - 4.48 eV

P, Torr	T, K							
	2600	2800	3000	3200	3400	3600	3800	4000
0.1	$3.3 \cdot 10^3$ (0.03)	170 (0.64)	14 (8.8)	3.3 (60)	3.1 (98)	3.1 (100)	3.1 (100)	3.2 (100)
0.3	$5.6 \cdot 10^3$ (0.02)	290 (0.37)	23 (5.2)	4.0 (42)	3.1 (95)	3.1 (100)	3.1 (100)	3.2 (100)
1	$1.0 \cdot 10^4$ (0.01)	530 (0.20)	41 (2.9)	5.6 (26)	3.1 (87)	3.1 (100)	3.1 (100)	3.2 (100)
3	$1.7 \cdot 10^4$ (0.006)	920 (0.12)	7.1 (1.7)	8.8 (16)	3.2 (72)	3.1 (99)	3.1 (100)	3.2 (100)
10	$3.2 \cdot 10^4$ (0.003)	$1.7 10^3$ (0.06)	130 (0.91)	1.5 (9.0)	3.6 (52)	3.1 (96)	3.1 (100)	3.2 (100)

In the case of three-atomic molecules, whose different degrees of freedom are excited equally, the dissociation efficiency is lower. This is conditioned by the necessity to excite more degrees of freedom, which consumes higher energy. Hence the ratio of temperature at which a noticeable dissociation proceeds, to dissociation energy in the case of three-atomic

molecules, when they decompose to diatomic ones, is markedly higher than in
the event of dissociation of diatomic molecules. For instance, for the pro-
cess of dissociation of molecule CO_2:

$$CO_2 \rightleftarrows CO + \frac{1}{2} O_2 - 2.9 eV$$

the maximum dissociation efficiency while heating under equlibrium conditi-
ons is 40% [24].

Table 6

The energy cost of formation, eV, and concentration of atom,
% (in parentheses) for heated nitrogen under conditions of
thermodynamic equilibrium $N_2 \rightleftarrows 2N - 9.77$ eV

P, Torr	T, K								
	5200	5400	5600	5800	6000	6200	6400	6600	6800
0.1	620 (0.32)	120 (1.8)	27 (8.5)	9.6 (32)	6.4 (67)	6.2 (97)	6.3 (100)	6.3 (100)	6.4 (100)
0.3	1100 (0.19)	210 (1.0)	45 (5.0)	13 (20)	6.9 (58)	6.3 (92)	6.3 (99)	6.3 (100)	6.4 (100)
1.0	1900 (0.10)	370 (0.57)	80 (2.8)	21 (11)	8.5 (38)	6.4 (80)	6.3 (97)	6.3 (100)	6.4 (100)
3.0	3400 (0.06)	640 (0.33)	140 (1.6)	35 (6.8)	10.6 (25)	6.7 (63)	6.3 (93)	6.3 (99)	6.4 (100)
10	6100 (0.03)	1200 (0.18)	250 (0.88)	60 (3.8)	18 (14)	8.1 (43)	6.4 (82)	6.3 (97)	6.4 (100)

In this case it is clear that in heating a gas under non-equilibrium
conditions one can attain a higher efficiency of dissociation, for then the
input energy is expended in not all degrees of freedom. Since the molecule
dissociation is accomplished through a vibrational excitation of molecule,
the effective dissociation can take place under non-equilibrium conditions,
in which the population of vibrationally excited states is higher than un-
der equilibrium conditions. Therefore, the molecule dissociation was sug-
gested to be performed in a non-equilibrium plasma with a high vibrational
temperature [13, 25-28].

Let us consider the manner of heating a carbon dioxide under non-equi-
librium conditions, when an rotational and translational temperatures fixed
at low level and only the vibrational temperature increases, which is the
some for various vibrational degrees of freedom. Under optimum conditions
(pressure - 1 atm, finite temperature - 3200 K) we obtain a maximum effici-
ency of dissociation, being equal, according to calculations [29], to 61%.
This seems to be a maximum figure for the dissociation efficiency. However,
experiments [28,31] show that the efficiency of dissociation over 80%, is
observed in the UHF - discharge. Let us analyze these data.

In a non-equilibrium gas the dissociation efficiency is growing for
two reasons. Firstly, a relatively less energy is used in an excitation of
translational and rotational degrees of freedom. Secondly, because of an-
harmonicity of molecules, the relative number of vibrationally - excited
molecules increases. Therefore, the same degree of dissociation is attained

at a lower vibtational temperature than at the absence of anharmonicity. However, since in an equilibrium gas of diatomic molecules the efficiency

Table 7

The energy cost of formation, eV, and concentration of oxygen atoms, % (in parentheses) for heated oxygen under conditions of thermodynamic equilibrium $O_2 \rightleftarrows 2O - 5.12$ eV

P, Torr	T, K						
	2600	2800	3000	3200	3400	3600	3800
0.1	7500 (0.013)	250 (0.43)	14 (8.6)	3.4 (70)	3.3 (99)	3.3 (100)	3.4 (100)
0.3	$1.3 \cdot 10^4$ (0.007)	430 (0.25)	24 (5.0)	3.8 (52)	3.3 (98)	3.3 (100)	3.4 (100)
1	$2.4 \cdot 10^4$ (0.004)	800 (0.14)	42 (2.8)	4.9 (33)	3.3 (95)	3.3 (100)	3.4 (100)
3	$4.1 \cdot 10^4$ (0.002)	1400 (0.08)	72 (1.6)	7.0 (21)	3.3 (88)	3.3 (100)	3.4 (100)
10	$7.5 \cdot 10^4$ (0.001)	2500 (0.04)	130 (0.89)	11 (12)	3.4 (72)	3.3 (99)	3.4 (100)

of their dissociation proves to be high, the study of this value variation in the non-equilibrium diatomic gas loses its practical interest.

The non-equilibrium has a significant effect on efficiency of dissocia-tion of three-atomic molecules. We shall further consider the most comple-tely studied case of dissociation of carbon dioxide molecules:

$$CO_2 \rightleftarrows CO + \frac{1}{2}O_2 - 2.9 eV$$
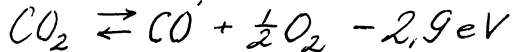
The maximum efficiency of this process amounts to 40% [24] in the case of thermodynamic equilibrium. In a non-equilibrium gas, when vibrational tem-perature exceeds considerably the translational and rotational ones, under optimal conditions (P=1 bar T = 3200 K), the maximum dissociation efficiency comes up to 61 % [29]. It was assumed in calculation [29] that an equilibrium is take place between various vubrational degrees of free-dom for excited states. Measurements [28] within the pressure range of 100 Torr at a gas excitation by an UHF-discharge result in an optimum dissoci-ation efficiency exceeding 80%. The authors explain such a high efficiency as a result of an excitation of one vibrational mode. But this assumption seems to us to be incorrect for two reasons. First, an excitation up to the states leading to dissociations, takes place as a result of many conse-cutive transitions between molecule vibrational states. Separate vibratio-nal states are unlikely to be separated so that in this case an exchange between different vibrational degrees of freedom will happen. Second, the mentioned assumption requires for that initially all input energy be con-tributed to one dergee of freedom. Direct experiments [32] show that under optimum conditions (electron mean energy - 1 - 1.5 eV), about 65% is con-tributed to an antisymmetric vibrational mode. Therefore, the efficiency of carbon dioxide dissociation calls for an additional analysis.

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