# INFLUENCE OF PLASMA PARAMETERS ON FORMATION RATE OF FULLERENES, NANOTUBES AND NANOPARTICLES

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**Introduction**

The model of fullerene formation proposed earlier [1,2,3] with taking into account carbon cluster charges was improved in this work. In the improved model for every carbon cluster $C_n$ of n atoms the distribution of concentration $c_n(l_n, T_n)$ was calculated depending on quantity of unsaturated bonds $l_n$ and vibration temperature $T_n$. It allowed to account carbon cluster cooling by buffer gas in classical approach, and also to account cooling and heating of clusters at bond breaking and formation.

**Results and discussion**

In development of improved model all carbon clusters were interpreted in the framework of reduced description first proposed in [4,5]. The reduced description scheme allows distinguishing clusters of different shape because of different quantity of radical bonds in them, and tracing cluster transformation processes. These processes lead to decrease of radical bonds quantity in cluster annealing process. At the same time this scheme is simple enough for intensive calculations of cluster kinetics.

In this scheme carbon cluster distribution $c(n,l,T,t)$ is the function of four parameters: time t, number of atoms n, number of radical (uncoupled) bonds l in cluster, and vibration temperature T.

At condensation in plasma carbon forms clusters with sp or $sp^2$ hybridization of atoms: chains, rings, parts of graphene planes and their combinations. It is obvious that fullerene molecules have l=0 radical bonds, monocyclic rings have one radical bond per atom (l=n), chains also have one radical bond per atom plus one radical bond per end atom (l=n+2). All other clusters (bicyclic, tricyclic clusters, incomplete fullerene fragments) have intermediate quantity of radical bonds per atom.

In the model it was taken into account that at carbon cluster annealing in buffer gas quantity of its radical bonds can decrease or increase due to formation or breaking of internal bonds, respectively. Formation of every new bond in cluster or between clusters released energy of 4.5-6 eV depending on C-C bond type. This energy redistributes between all atoms of cluster due to vibration nonlinearity, and sufficiently increases vibration temperature T of cluster.

In the model carbon cluster cooling by buffer gas was taken into account in classical approach. If vibration temperature T of cluster is higher than buffer gas temperature $T_{gas}$, it is cooled by collisions with gas atoms. Otherwise, at $T < T_{gas}$ cluster is heated by buffer gas. At the collisions with buffer gas atoms also the translational energy of carbon cluster $T_{kin}$ accounted. Accounting the cooling of clusters by buffer gas during their coagulation was developed for the first time.

Solution of kinetic equations for concentrations of all cluster isomers (about 4000) having different temperature and number of radicals need large amount of operations. Therefore the following simplifications were accepted.

In calculations only clusters with even number of atoms were taken into account, because mass-spectroscopic measurements at carbon evaporation had shown that amount of clusters with even number of atoms essentially exceed amount of clusters with odd number of atoms [6].

Furthermore, the calculations were carried out for infinitely small volume cell, so the cluster distribution $c(n,l,T,t)$ did not included dependence on spatial coordinates. Also, for conservation of solution stability at big quantity of different clusters, plasma temperature and electron concentration were fixed in calculation. It allowed carrying out calculation of fullerene formation for different relations of plasma parameters, which can be achieved by injection of easy-ionized admixtures into plasma.

According to [7,8,9] and our recent spectral investigations of fullerene high-frequency (HF) arc at atmospheric pressure (Fig.1), the initial stage of cluster formation corresponds to high concentration of $C_2$ and absence of other clusters. Therefore, at the start of calculation (t=0) the only clusters were $C_2$ dimers, i.e. at the start concentration $c(n, l, T, t) \neq 0$ only for n=2 and l=4.
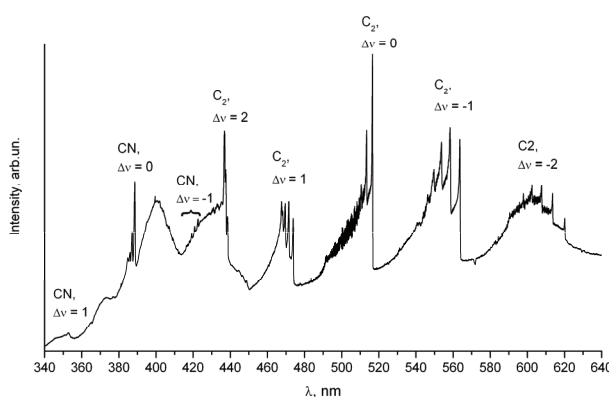
Fig. 1. Radiation spectrum of temperature zone 3000–4000K of fullerene plasma of HF arc at atmospheric pressure in 340-640 nm range.
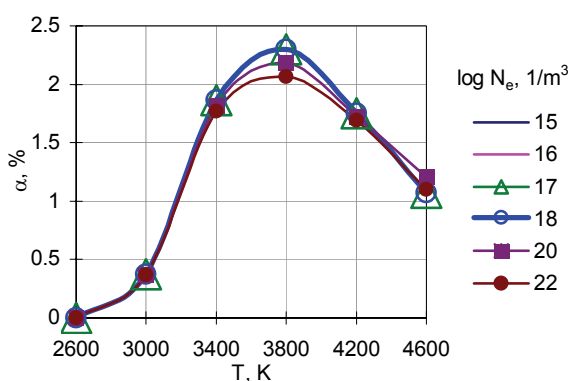


Fig.2. Dependence of calculated fullerene yield on temperature and electron concentration.

In the Fig.2 temperature dependencies of fullerene yield ($C_{60}+C_{70}$) for model time $5 \cdot 10^{-5}$ s for different electron concentrations are shown. Maximum fullerene yield obtained at buffer gas temperature 3800K, because at lower temperatures probability of isomerization barriers decreases, and at higher temperatures probability of bond breaking increases. The obtained value is in agreement with estimations of temperature of fullerene formation about 3000-3500K. Influence of electron concentration is notable only at maximum fullerene yield and do not exceed 10% of the yield.

**Conclusions**

As a result of model and real experiment realization the more accurate fullerene formation rates were obtained, and it was shown that for the HF arc of atmospheric pressure, as for DC arc at 100 torr, dimer $C_2$ is the main carbon cluster presented at initial stage of fullerene plasma corresponding to temperature 3000 – 4000 K.

**References**
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