Two-Dimensional Modeling of Positive Streamer Propagation in Flue Gases in Sphere-Plane Gaps

Natalya Yu. Babaeva and George V. Naidis

***Abstract*— The results of two-dimensional numerical simulation of positive streamer dynamics in flue gases in sphere-plane electrode configuration are presented. The *G*-values (numbers of radicals produced per 100 eV of input electrical energy) for the production of chemically active particles participating in flue gas cleaning from toxic components are calculated. Obtained *G*-values are almost independent on the discharge conditions. Simple estimates for *G*-values, based on analytical streamer theory, are shown to agree with the results of numerical simulation.**

***Index Terms*— Corona discharge, flue gas cleaning, positive streamer.**

I. Introduction

PULSED positive corona discharges in flue gases are actively studied in connection with their use for gas cleaning from toxic components NO_x and CO_2 [1]. Such discharges have a structure of a number of streamers—thin plasma channels propagating in discharge gap. Active particles taking part in the removal of toxic components are produced in the regions of high electric field—in the streamer heads. The problem of simulation of the cleaning process includes two stages. The first stage is calculation of the rates of primary active particles production by streamers, the second is the modeling of subsequent chemical transformations in the flue gas mixture. The chemical part of the problem has been considered in a number of works [2]–[8]. In most of these works, some assumptions are made about initial concentrations of radicals, because available information about the efficiency of generation of active particles by streamers is rather poor. In previous works [9]–[11], calculations of radical production by streamers in flue gases have been done with the use of one-dimensional (1-D) streamer models. In the frame of 1-D models, the results depend on the choice of the value of streamer radius which is an input parameter of the model. A more rigorous approach is based on two-dimensional (2-D) streamer modeling. In the present paper, the results of 2-D simulation of positive streamer propagation in flue gas are given. The G-values (numbers of particles produced per 100 eV of input electrical energy) for the production of primary chemically active components are obtained.

Manuscript received February 24, 1997; revised September 19, 1997. This work was supported by INTAS under Contract 94-4207 and by NWO (Dutch Organization for Scientific Research).

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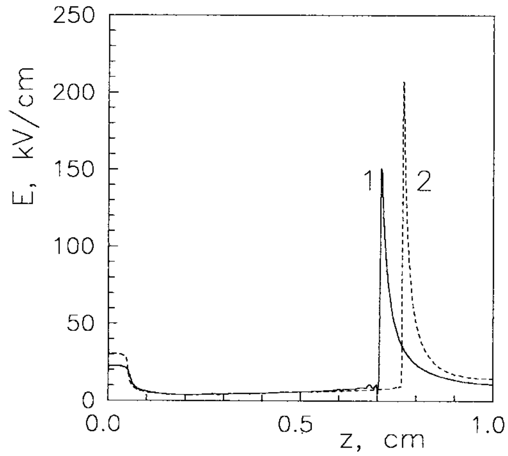
Publisher Item Identifier S 0093-3813(98)00695-X.

II. Simulations

The 2-D streamer model is analogous to that used earlier for air, and is described in detail in [12] and [13]. Streamer propagation in the sphere-plane gap (the sphere radius $R_{sph} = 0.05$–0.2 cm, the ratio of the gap length d to the sphere radius $d/R_{sph} = 5$–20) in the mixture $N_2 : O_2 : CO_2 : H_2O = 0.71 : 0.05 : 0.08 : 0.16$ at the molecule number density $n = 2.5 \cdot 10^{19}$ cm^{-3} and the gas temperature $T = 340$ K has been simulated. As it has been shown in [9], the electron energy distribution function (EEDF) in flue gas mixtures is close (at the values of reduced electric field E/n typical for streamer propagation) to the EEDF in air. So in our model for flue gases, the rate constants of excitation and dissociation of gas components have been taken calculated with EEDF in air. Approximations of the excitation rate constants of nitrogen triplet electronic states and the rate constants of oxygen dissociation and dissociative excitation presented in [14] have been used. The data on the nitrogen dissociation rate constant have been taken from [15]. Calculation of carbon dioxide dissociation and water dissociative attachment rate constants has been made with the use of cross sections presented in [16] and [17]. The dependencies of the ionization coefficient and the drift velocity of electrons on E/n in flue gas have been taken the same as in air, in accordance with [9]. The photoionization model derived in [18] for nitrogen-oxygen mixtures has been used modified by the additional account of absorption of ionizing photons by H_2O and CO_2 molecules (corresponding absorption coefficients are taken from [19] and [20]). The attachment coefficient in flue gas differs from that in air, but the role of attachment is not essential for short streamers considered here (see [12]). The difference between parameters of streamers in air and flue gases is caused mainly by the shortening of the path length of ionizing photons in flue gases in comparison with air due to the strong absorption by H_2O and CO_2 molecules.

In Fig. 1, the distributions of the electric field E and the electron number density n_e along the streamer axis z are compared for streamers propagating in flue gas and in air. The value of the electric field in the streamer head E_h in flue gas is greater than in air. Correspondingly, the electron number density in the streamer channel n_c in flue gas is also greater than in air (correlation of E_h and n_c obtained by 2-D simulation agrees with the results of analytical streamer theory; see [12]).

The dependencies of the velocity of streamer propagation V and current I on streamer length L in flue gas and in air are shown in Fig. 2. The values of V and I in flue gas are



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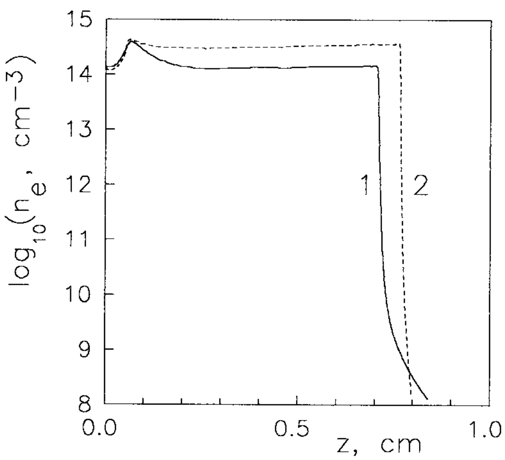
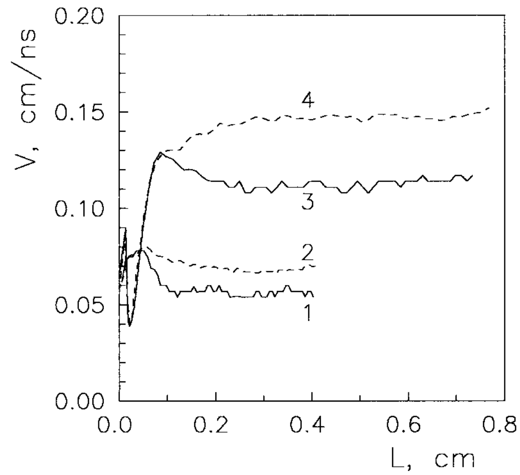


(b)

Fig. 1. (a) Electric field and (b) electron number density distributions along the streamer axis in air (lines 1) and flue gas (lines 2), for sphere radius $R_{sph} = 0.2$ cm, gap length $d = 1$ cm, applied voltage $U = 14$ kV.



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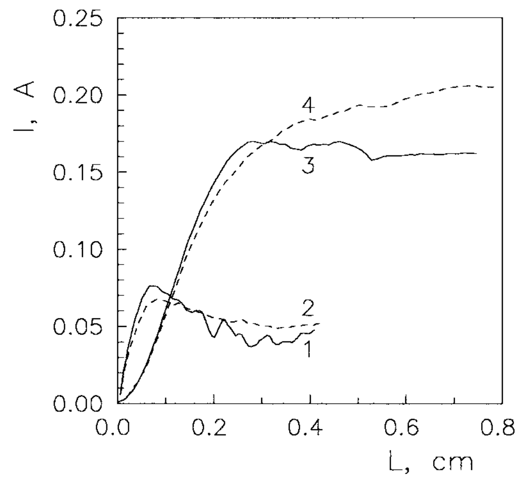


(b)

Fig. 2. (a) Dependencies of streamer velocity and (b) current on its length in air (lines 1, 3) and flue gas (lines 2, 4), for $R_{sph} = 0.05$ cm, $d = 0.5$ cm, $U = 6$ kV (lines 1, 2) and for $R_{sph} = 0.2$ cm, $d = 1$ cm, $U = 14$ kV (lines 3, 4).

slightly greater than in air (for the same external conditions). Streamer radius R in flue gas is about 10–20% less than in air.

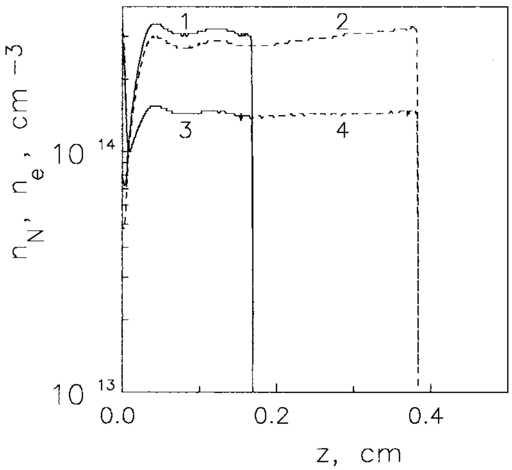
The results of simulation show that, as for streamers in air [12], the values of E_h and n_c are almost independent on the external conditions of the discharge (the applied voltage, the sphere radius, and the gap length). Streamer radius, velocity, and current increase with the applied voltage.

Concentrations have been calculated of chemically active components generated in the flue gas mixture: electronically excited nitrogen molecules N_2^* (producing radicals in collisions with O_2, CO_2, and H_2O molecules), nitrogen atoms N, oxygen atoms in ground state $O(^3P)$, and in excited state $O(^1D)$, CO and OH radicals. In Fig. 3, axial values of concentrations n_e, n_N and linear number densities N_e, N_N (concentrations integrated in radial direction) of electrons and nitrogen atoms in streamer channel are presented for two time moments of streamer propagation in flue gas. It is seen that atoms are generated mainly in streamer head, their concentration in the channel does not change during streamer propagation. Concentration of electrons in the channel slightly decreases with time due to three-body attachment to oxygen molecules. The ratio n_e/n_N is greater than N_e/N_N. It means that the effective width of the radial distribution of electrons [which can be estimated as $(4N_e/\pi n_e)^{1/2}$] is smaller than that of nitrogen atoms. This fact is caused by stronger dependence of ionization coefficient on E/n in comparison with the dissociation coefficient (the electric field in the streamer head is maximal at the axis and decreases in radial direction).

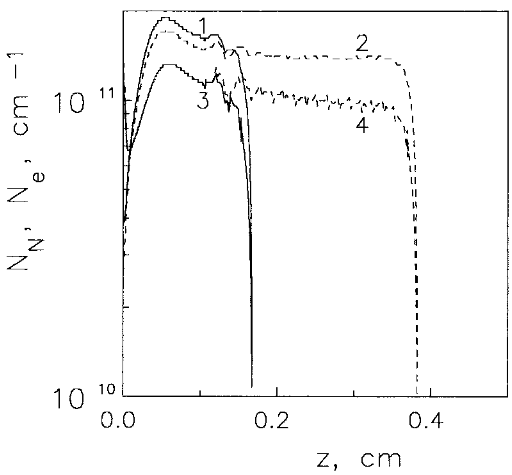
The results of calculation show that, as in air [21], the dependence of the G-values on external conditions in flue gas is weak. As an example, in Fig. 4, the G-values for nitrogen atoms production are given corresponding to various parameters of the gap and applied voltages. They slowly decrease with growth of streamer length. Calculated G-values for production of chemically active components are given in Table I. Also, estimations of the G-values are presented in the table obtained with the use of analytical streamer theory (see below).

III. Discussion

It is interesting to compare the G-values obtained by 2-D modeling and estimated with the use of simple analytical



(a)



(b)

Fig. 3. (a) Distributions of concentrations and (b) linear number densities of electrons (lines 1, 2) and nitrogen atoms (lines 3, 4) along the streamer axis, for $R_{sph} = 0.05$ cm, $d = 0.5$ cm, $U = 6$ kV, at two time moments t: 2.4 ns (lines 1, 3) and 5.6 ns (lines 2, 4).

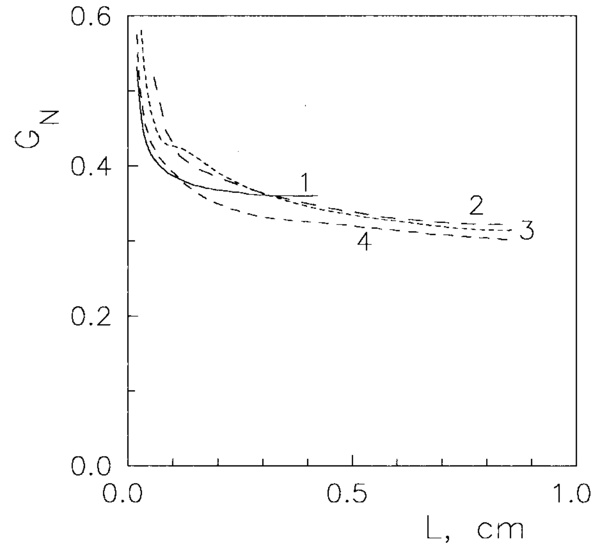


Fig. 4. Dependence of the G-factor for nitrogen atoms production on streamer length in flue gas, for $R_{sph} = 0.05$ cm, $d = 0.5$ cm, $U = 6$ kV (line 1); $R_{sph} = 0.2$ cm, $d = 1$ cm, $U = 20$ kV (line 2) and 14 kV (line 3); $R_{sph} = 0.1$ cm, $d = 1$ cm, $U = 8$ kV (line 4).

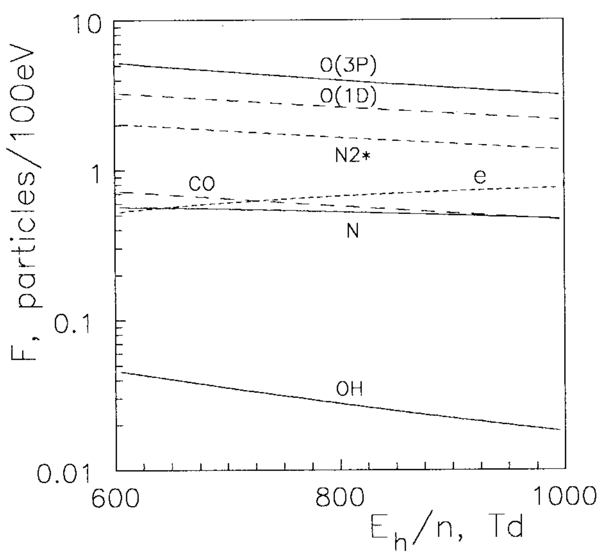


Fig. 5. Dependencies of the values F for the production of active particles in flue gas mixtures on the reduced electric field in streamer head (see the text).

relations [21] based on the analytical streamer theory [22]. The value G_j corresponding to the production of active particles of sort j in reactions with high enough energy thresholds (when particles are generated mainly in streamer head) is given by the expression

$$G_j = \frac{C}{\ln(L/R)} \sum x_i F_{ij} \tag{1}$$

where C is the numerical factor of the order of 1, x_i is the relative concentration of molecules of sort i in the mixture, and the values F_{ij} are

$$F_{ij} = \frac{\displaystyle\int_{E_c/n}^{E_h/n} \frac{K_{ij}}{V_{dr}}\, d(E/n)}{e(E_h/n)^2}. \tag{2}$$

Here K_{ij} is the reaction rate constant for the production of particles j in collisions of electrons with molecules i, V_{dr} is the drift velocity of electrons. The upper limit of the integral in (2) is the maximal value of the reduced electric field in the streamer head E_h/n, the lower limit is the reduced field in the streamer channel E_c/n (note that the dependence of the integral on the lower limit is very weak).

In Fig. 5, the values F for the production of active particles in flue gas mixtures (calculated with the use of EEDF for air) are presented as functions of the E_h/n value. Lines marked "O(3P)," "O(1-D)," "CO," "N," "N2*," and "OH" correspond to generation of these particles at dissociation and dissociative excitation of O_2 molecules, dissociation of CO_2 and N_2 molecules, excitation of triplet states of N_2, and dissociative attachment to H_2O, respectively. At calculating G-factors with the use of (1), these F-values must be multiplied by corresponding concentrations of molecules in the mixture. The

TABLE I
G-VALUES IN THE MIXTURE
$N_2 : O_2 : CO_2 : H_2O = 0.71 : 0.05 : 0.08 : 0.16$

	G_N	G_{OH}	G_{CO}	$G_{N_2^*}$	$G_{O(^3P)}$	$G_{O(^1D)}$
2D	0.30-0.38	0.007-0.011	0.05-0.06	1.2-1.5	0.28-0.36	0.14-0.19
(1)	0.37	0.007	0.045	1.2	0.25	0.13

line marked "e" in Fig. 5 describes generation of electrons and takes into account all ionization channels in the flue gas mixture (it is assumed that the ionization rate in flue gases is the same as in air; see above). Note that in the range of E_h/n, typical for streamer propagation in flue gases (800–900 Td), the dependencies of F-values on E_h/n are rather weak. So the inaccuracy at calculation of F-values related with the use of EEDF in air instead of real EEDF in the flue gas mixture is small enough, and the F-values given in Fig. 5 can be used for flue mixtures with various relative concentrations of main components.

The G-values estimated in accordance with (1) for the considered mixture $N_2 : O_2 : CO_2 : H_2O = 0.71 : 0.05 : 0.08 : 0.16$ are presented in Table I (for $E_h/n = 800$ Td and the ratio $C/\ln(L/R)$ in (1) taken equal to 1). The G-value for oxygen atoms $O(^3P)$ includes deposits of both O_2 and CO_2 dissociation. Comparison of these estimates with the results of 2-D simulation shows their reasonable agreement.

For the modeling of chemical transformations in the mixture after streamer discharge, the knowledge of not only G-factors but also of the concentrations of radicals in the streamer trail is needed. Their values can be easily estimated by comparison of corresponding G-factors with the G-factor for generation of electrons $(G_e \approx 0.7)$ taking into account that the concentration of electrons in streamer channel is $\sim (3–4) \cdot 10^{14}$ cm^{-3}. Thus, for the considered flue gas mixture, such an estimation gives concentration of nitrogen atoms $\sim (1.6–2.1) \cdot 10^{14}$ cm^{-3} close to the result of 2-D simulation (see Fig. 3).

Note that the model used in this work does not take into account the streamer–cathode interaction. So it cannot describe the stage when the streamer head approaches the cathode and, respectively, the later stage of the electric field redistribution inside the channel after crossing the gap.

Due to numerical limitations, the simulation has been made of streamers in relatively short gaps. In the case of longer gaps, used in practice, situation will not change till the time of streamer propagation is less than the characteristic time τ_c of decrease of the channel conductivity (at atmospheric molecule number densities τ_c is several tens of nanoseconds). So, the results obtained in this work can be applied in conditions when the duration of the voltage pulse is shorter than τ_c and than the time needed for streamer to cross the gap.

IV. SUMMARY

Numerical simulation of positive streamer propagation in flue gas shows that, analogously to streamers in air [12], some of the streamer parameters (radius, velocity, current) essentially depend on the external conditions of the discharge, while other parameters (the electric field in streamer head, electron and radical concentrations in streamer channel, the G-values for radical production) are almost constant. The G-values in a given flue gas mixture can be estimated with the use of a simple analytical expression (1).

ACKNOWLEDGMENT

The authors would like to thank Dr. E. M. van Veldhuizen and Prof. W. R. Rutgers for helpful discussions.

REFERENCES

[1] *Non-Thermal Plasma Techniques for Pollution Control*, B. M. Penetrante and S. E. Schultheis, Eds. Berlin: Springer, 1993.
[2] B. M. Penetrante, "Plasma chemistry and power consumption in nonthermal DeNO$_x$," in *Non-Thermal Plasma Techniques for Pollution Control*, B. M. Penetrante and S. E. Schultheis, Eds. Berlin, Germany: Springer, 1993, pt. A, pp. 65–89.
[3] R. P. Dahiya, S. K. Mishra, and A. Veefkind, "Plasma chemical investigations for NO_x and SO_2 removal from flue gases," *IEEE Trans. Plasma Sci.*, vol. 21, pp. 346–348, 1993.
[4] G. Yu. Alekseev, A. V. Levchenko, and V. A. Bityurin, "Flue gas cleaning by pulse corona: Chemical kinetics and heat- and masstransfer in NO/SO_2 removal," Res. Rep. 93/2, IVTAN-ANRA, Moscow, U.S.S.R., 1993.
[5] J. J. Lowke and R. Morrow, "Theoretical analysis of removal of oxides of sulphur and nitrogen in pulsed operation of electrostatic precipitators," *IEEE Trans. Plasma Sci.*, vol. 23, pp. 661–671, 1995.
[6] W. Sun, B. Pashaie, S. Dhali, and F. I. Honea, "Non-thermal plasma remediation of SO_2/NO using a dielectric-barrier discharge," *J. Appl. Phys.*, vol. 79, pp. 3438–3444, 1996.
[7] A. C. Gentile and M. J. Kushner, "Microstreamer dynamics during plasma remediation of NO using atmospheric pressure dielectric barrier discharges," *J. Appl. Phys.*, vol. 79, pp. 3877–3885, 1996.
[8] M. B. Zheleznyak and E. A. Filimonova, "Modeling of gas phase chemical reactor based on pulsed streamer discharge for removal of toxic admixtures," *High Temp.*, vol. 36, no. 3, 1998.
[9] I. Gallimberti, "Impulse corona simulation for flue gas treatment," *Pure Appl. Chem.*, vol. 60, pp. 663–674, 1988.
[10] A. A. Kulikovsky, A. Kh. Mnatsakanian, G. V. Naidis, and Yu. M. Solozobov, "Models for positive corona streamer propagation in air and combustion products," in *Proc. XXI Int. Conf. Phenom. Ioniz. Gases*, Bochum, Germany, 1993, vol. 1, pp. 297–298.
[11] R. Morrow, J. J. Lowke, A. J. Paulson, and A. J. Prokopiuk, "Pulsed positive corona to reduce NO_2 and SO_2 emissions," in *Proc. XXII Int. Conf. Phenom. Ioniz. Gases*, Hoboken, NJ, 1995, vol. 1, pp. 143–144.
[12] N. Yu. Babaeva, and G. V. Naidis, "Two-dimensional modeling of positive streamer dynamics in nonuniform electric fields in air," *J. Phys. D: Appl. Phys.*, vol. 29, pp. 2423–2431, 1996.
[13] ——, "Dynamics of positive and negative streamers in air in weak uniform electric fields," *IEEE Trans. Plasma Sci.*, vol. 25, pp. 375–279, 1997.
[14] N. L. Aleksandrov, A. E. Bazelyan, E. M. Bazelyan, and I. V. Kochetov, "Modeling of long streamers in gases at atmospheric pressure," *Plasma Phys. Rep.*, vol. 21, pp. 57–69, 1995.
[15] B. M. Penetrante, M. C. Hsiao, B. T. Merritt, G. E. Vogtlin, and P. H. Wallman, "Comparison of electrical discharge techniques for nonthermal plasma processing of NO in N_2," *IEEE Trans. Plasma Sci.*, vol. 23, pp. 679–687, 1995.
[16] D. I. Slovetsky, *Mechanisms of Chemical Reactions in Nonequilibrium Plasmas*. Moscow: Nauka, 1980 (in Russian).
[17] R. N. Compton and L. G. Christophorou, "Negative ion formation in H_2O and D_2O," *Phys. Rev.*, vol. 154, pp. 110–121, 1967.
[18] M. B. Zheleznyak, A. Kh. Mnatsakanian, and S. V. Sizykh, "Photoionization of nitrogen-oxygen mixtures by gas discharge radiation," *High Temp.*, vol. 20, pp. 357–361, 1982.
[19] R. D. Hudson, "Critical review of ultraviolet photoabsorption cross sections for molecules of astrophysical and aeronomical interest," *Rev. Geophys. Space Phys.*, vol. 9, pp. 305–406, 1971.
[20] G. R. Cook, P. H. Metzger, and M. Ogava, "Absorption, photoionization and fluorescence of CO_2," *J. Chem. Phys.*, vol. 44, pp. 2935–2944, 1966.
[21] G. V. Naidis, "Modeling of plasma chemical processes in pulsed corona discharges," *J. Phys. D: Appl. Phys.*, vol. 30, pp. 1214–1218, 1997.
[22] M. I. Dyakonov and V. Yu. Kachorovsky, "Theory of streamer discharge in semiconductors," *Sov. Phys. JETP*, vol. 67, pp. 1049–1054, 1988.

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