## PULSE-PERIODIC DISCHARGE IN A PLASMACHEMICAL FERROELECTRIC PACKED-BED REACTOR

S. A. Zhdanok, G. M. Vasil'ev, and V. A. Vasetskii

A plasmachemical method of cleaning the exhaust gases of Diesel engines from toxic components in a ferroelectric packed-bed reactor is described. Special features of the electric circuit of this plasmachemical system and the energy efficiency of the process of cleaning under the conditions of pulse-periodic input of energy to the discharge are considered.

For realization of different types of electric discharges in electrophysical devices intended for reduction of the toxicity of exhaust gases (EG) the following basic configurations of electrode systems are used: point-plane [1–3], thin wire in tube [4, 5], thin wire-plane [6, 7], tube in tube [8, 9], and packed-bed reactors [8, 10, 11]. In practice, each of the presented electrode systems can have a wide variety of designs and several basic variants can be used in a specific device simultaneously.

One variant of improving the volume characteristics and efficiency of a plasmachemical system is the use of pulse discharge in the ferroelectric bed (BaTiO<sub>3</sub>, PbTiO<sub>3</sub>, PbZrO<sub>3</sub>, etc.). Dielectric granules are kept in a chamber made of an insulating material (quartz glass, ceramics) by two mesh electrodes to which high voltage is applied. In this case, a strong electric field forms near each dielectric element, as a result of which high-energy electrons appear within the entire volume of the discharge chamber. Ferroelectrics strengthen the electric field in the contact regions between neighboring elements, thus leading to microdischarges in the gaps. A high value of the dielectric constant increases the effect of plasma formation and production of active radicals in the volume of the discharge chamber of the reactor [10, 11].

Special Features of the Electric Circuit of the Packed-Bed Reactor. The ferroelectric packed-bed reactor can be presented in the form of a large number of parallel chains with each chain consisting of separate condensers  $(C_{i,p} \text{ and } C_{i,g})$  and resistors  $(R_{i,p} \text{ and } R_{i,g})$ .

An equivalent electric circuit that involves the total structural inductance is given in Fig. 1a. If the applied voltage is small (microdischarges between particles are absent), then the main role is played by  $C_{i,p}$  and  $C_{i,g}$  since the resistances  $R_{i,p}$  and  $R_{i,g}$  tend to infinity. With microdischarges between the particles the capacitances  $C_{i,g}$  are by-passed by the resistances of the discharges  $R_{i,g}$  ( $R_{i,g} \sim 1-10^2 \Omega$ ), and the resistance  $R_{i,p}$  remains much higher. For this case, an equivalent electric circuit can be represented by the combination of  $C_{i,g}$  and  $R_{i,g}$  (Fig. 1b).

On introduction of the total resistance R and the capacitance C of the layer, the equivalent electric circuit can be modified (Fig. 2). It should be noted that the schematic diagram of the ferroelectric packed-bed reactor (Fig. 2) in electric discharge differs from the systems of reactors of the wire–plate or point–plate types, where corona streamer discharge is realized. In equivalent electric circuits of the reactors of the wire–plate or point–plate types, the equivalent R and C are connected in parallel.

Parameters of the Packed-Bed Reactor. We studied a reactor with the following parameters:

Maximum size of spherical particles (PbTiO <sub>3</sub> )	4.5 mm
Permittivity of particles	400-500
Mean porosity of the bed	36%
Interelectrode distance	40 mm

A. V. Luikov Heat and Mass Transfer Institute, National Academy of Sciences of Belarus, 15 P. Brovka Str., Minsk, 220072, Belarus. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 78, No. 1, pp. 174–177, January–February, 2005. Original article submitted October 14, 2004.

UDC 536.46



Fig. 1. An equivalent electric circuit of the ferroelectric packed-bed reactor without (a) and with (b) discharge.



Fig. 2. An equivalent electric circuit of the ferroelectric packed-bed reactor in electric discharge.



Fig. 3. The scheme of measurement of the capacitance of the ferroelectric packed-bed reactor in electric discharge: C, reactor capacitance; R, active resistance of the reactor;  $L_c$ , control inductance; G, discharge gap;  $R_{lim}$ , current-limiting resistor;  $R_d$ , voltage divider; DC, power sources; HV, high voltage.

*The capacity of the packed-bed reactor* was measured by a precise digital LCR-meter and was  $73 \cdot 10^{-12}$  F; the capacity of the reactor without particles was  $15 \cdot 10^{-12}$  F.

The capacity of the ferroelectric packed-bed reactor with an electric discharge was determined according to the following scheme (Fig. 3).

The intrinsic inductance of the reactor  $L \ll L_c = 0.24 \cdot 10^{-6}$  H. The measurements were conducted by the following scheme: the condenser C was charged via the current-limiting resistor  $R_{\rm lim}$  till a certain voltage U, upon reaching of which the discharge gap G was broken down and harmonic oscillations appeared in the  $C-L_c$  circuit. These oscillations were recorded by an oscillograph. Since the value of  $L_c$  is rather high and the amplitude of oscillations changes slightly, the condition  $R \ll 2(L_c/C)^{1/2}$  holds and the value of the capacity C can be calculated by the formula  $T = 2\pi (L_c C)^{1/2} \rightarrow C = T^2/(4\pi^2 L_c)$ , where T is the period of the first oscillation. According to the oscillogram,  $T = 1.2 \cdot 10^{-6}$  sec, whence  $C_{\rm exp} \cong 152 \cdot 10^{-12}$  F.

Theoretical calculation of the ferroelectric packed-bed reactor ( $\epsilon = 400-500$ ) showed that the capacity *C* strongly depends on the type of particle packing in the bed (see Table 1):

$$C = C_{\text{theor}} = S \varepsilon_{\text{ef}} \varepsilon_0 / H$$
,

where S is the surface area of the layer, H is the thickness of the particle layer, and  $\varepsilon = 8.85 \cdot 10^{-12}$  F/m is the dielectric constant.

	Type of particle packing			
Permittivity of particles	cubic		hexagonal	
	$C_{\text{theor}} \cdot 10^{-12}$ , F	$\epsilon_{eff}$	$C_{\text{theor}} \cdot 10^{-12}$ , F	$\epsilon_{eff}$
90	7.5	13.5	14.7	26.5
300	17.3	31.2	42.75	76.9
660	30.0	58.1	—	—
2000	87.1	156.7		

TABLE 1. Capacitance and Effective Permittivity of the Ferroelectric Bed



Fig. 4. The scheme of measurement of the inductance of the ferroelectric packed-bed reactor in electric discharge:  $L_c$ , R, and C, inductance, resistance, and capacitance; G, discharge gap;  $R_{lim}$ , current-limiting resistor; RgC, Rogovskii band.

The difference between  $C_{\text{theor}}$  and  $C_{\text{exp}}$  can be explained as follows: the particles used in the experiments had an irregular spherical shape and slightly differed in size. These factors decreased the number of contact points between the particles and, thus, the equivalent capacity of the bed and the number of microcharges.

The inductance of the ferroelectric packed-bed reactor in electric discharge was determined by the scheme shown in Fig. 4.

As in the previous case, the condenser C was charged through the current-limiting resistor  $R_{lim}$  till certain voltage U, upon reaching of which the discharge gap G was broken down and harmonic oscillations appeared in the  $C-L_c$  circuit. Since the amplitude of these oscillations decreases slightly (10–15%), the condition  $R \ll 2(L_c/C)^{1/2}$  holds and the value of the inductance L can be calculated using the expression  $L = T^2/(4\pi^2 C) = 1.7 \cdot 10^{-6}$  H, where the period of the first oscillation  $T = 10^{-7}$  sec.

It is difficult to accurately determine *L* from the expression  $T = 2\pi/(1/LC) - R^2(4L^2))^{1/2}$  because of the difficulties in the determination of *R*, which changes during the whole period of discharge. In this case, the impedance of the reactor can be estimated through measurements of *L* and *C*:  $|Z| = (L/C)^{1/2} \cong 990 \Omega$ . This value strongly differs from the results obtained in processing of oscillograms of current and voltage:  $|Z| \cong 70-80 \Omega$ .

The development of the forming line with such an impedance is not a problem. The contribution of energy to the discharge in the use of a simple reactor and the forming line is most effective when  $|Z_r| = |Z_{f,lin}|$ . During a short period of time (determined by the degree of concordance between the impedance of the pulse generator and the active resistance of the reactor) all energy is distributed between the capacity of the forming line and the capacity of the reactor (in proportion to their own capacities). In the experimental studies, the discharge is usually calculated from the oscillograms of current and voltage of the discharge: |Z| = U(t)/I(t). However, in order to determine the discharge resistance proper it is necessary to divide the total current by the current of discharge through R,  $I_R(t)$ , and the current through C,  $I_C(t)$ , in the case where R and C are connected in parallel (the wire–plate type reactor). When R and C are connected in series (the ferroelectric packed-bed reactor), voltages  $U_R(t)$  and  $U_C(t)$  must be separated. In both cases, this separation of parameters is difficult from the engineering point of view; therefore, complete agreement is virtually impossible.

Energy Characteristics of Pulse-Periodic Discharge in the Ferroelectric Packed Bed. Exhaust gases of a CN-6D Diesel engine (composition:  $O_2 = 18.5\%$ , CO = 500–550 ppm,  $CO_2 = 1.8\%$ ,  $NO_x = 200-250$  ppm,  $SO_2 = 0$ )



Fig. 5. Dependence of the efficiency of the decrease in the content of nitrogen oxides and exhaust gases on the value of the specific energy contribution: 1) f = 500; 21000; 31500 Hz. W, W·h/m<sup>3</sup>;  $\eta$ , %.

Fig. 6. Efficiency of cleaning of exhaust gases from nitrogen oxides at different pulse recurrence frequencies: 1) f = 1500; 2) 1000; 3) 500 Hz. Q, W/(1 ppm NO<sub>x</sub>); B, kV/cm.

that were preliminarily cleaned from soot were pumped through the reactor with  $PbTiO_3$  particles at a volume flow rate of 0.1–0.2 m<sup>2</sup>/h.

The results of the experiments on reduction of the concentration of nitrogen oxides in the plasma of particle discharge in the ferroelectric bed under the effect of high-voltage unipolar pulses of microsecond length are given in Fig. 5.

The content of nitrogen oxides in exhaust gases decreases monotonically as the contributed energy and pulse recurrence frequency increase. The energy efficiency of the process of cleaning can be characterized by the value of Q, which for our experiments is given in Fig. 6.

As is seen, the optimum strength of the electric field lies within the limits 3.5-4.5 kV/cm. For PbTiO<sub>3</sub> particles at 5-5.5 kV/cm the through breakdown of the discharge interelectrode gap filled by ferroelectric particles begins. At an initial content of NO<sub>x</sub> ~ 250 ppm and about a 60% decrease in the concentration of nitrogen oxides in exhaust gases, the specific energy contribution to the discharge is ~15 W·h/m<sup>3</sup>.

Thus, in the value of density of energy contributed to the gas this method is comparable with cleaning in plasma of the pulse corona discharge. Upon transition to cleaning of large volumes of gas, the gasdynamic resistance of the reactor, heating of the dielectric, and cost of the packing material begin to manifest themselves.

## NOTATION

*B*, reduced voltage, kV/cm; *C*, capacitance, F; *f*, frequency, Hz; *H*, thickness of the bed of particles, m; *I*, current, A; *L*, inductance, H; *Q*, specific energy, W/(1 ppm NO<sub>x</sub>); *R*, resistance,  $\Omega$ ; *S*, surface area of the bed, m<sup>2</sup>; *T*, period of oscillations, sec; *t*, time, sec; *U*, voltage, V; *W*, specific energy contribution, W·h/m<sup>3</sup>; *Z*, discharge impedance,  $\Omega$ ;  $\varepsilon$ , permittivity of particles;  $\varepsilon_0 = 8.85 \cdot 10^{-12}$ , dielectric constant, F/m;  $\eta$ , efficiency, %. Indices: g, discharge gap; i, individual particle; c, circuit; p, particle; lim, limiting; ef, effective; exp, experiment; theor, theoretical calculation; r, reactor; f.lin, forming line; d, divider.

## REFERENCES

- 1. A. Mizuno, J. S. Clements, and R. H. Davis, A method for the removal of sulphur dioxide from exhaust gas utilizing pulsed streamer corona for electron energization, *IEEE Trans. Ind. Appl.*, **IA–22**, May/June, No. 3, 516–521 (1986).
- Yu. S. Akishev, A. A. Deryugin, I. V. Kochetkov, A. P. Napartovich, and N. I. Trushkin, Experimental study of cleaning of flue gases from NO by a stationary glowing discharge at atmospheric pressure, in: *Proc. of Seminar* [in Russian], IVTAN, Moscow (1992), pp. 40–43.

- 3. M. C. Park, D. R. Chang, M. H. Woo, G. J. Nam, and S. P. Lee, Multi-stage pulsed corona discharge system for reduction of NO<sub>x</sub> emission from diesel engine vehicle (SAE 982514), in: *Plasma Exhaust Aftertreatment* (SP-1395) (1998), pp. 93–99.
- 4. J.-O. Chae, G. Vasiliev, S. M. Lee, Y. N. Jun, Y. S. Jun, and B. G. Kim, NO<sub>x</sub> removal with the pulse streamer corona discharge, in: *Proc. Korean Society of Environmental Engineers*, May 1993, Kung Hee University (1993), pp. 173–177.
- 5. G. M. Vasiliev, J.-O. Chae, S. C. Chung, Y. S. Jeong, and D. S. Han, NO<sub>x</sub> removal with the pulse streamer corona discharge, in: *Proc. Zel'dovich Memorial Conf.*, September 1994, Moscow, Russia (1994), p. 75.
- 6. L. G. Krishtopa and L. N. Krasnoperov, NO<sub>x</sub> destruction in air and nitrogen by dielectric barrier corona discharge (SAE 982510), in: *Plasma Exhaust Aftertreatment* (SP–1395) (1998), pp. 67–72.
- 7. S. A. Zhdanok, G. M. Vasil'ev, and V. A. Vasetskii, Study of decomposition of  $NO_x$  in nonequilibrium plasma, in: *Heat and Mass Transfer-93/94* [in Russian], Coll. of Sci. Papers of the Heat and Mass Transfer Institute, National Academy of Sciences of Belarus, Minsk (1994), pp. 73–76.
- 8. G. M. Vasiliev, S. A. Zhdanok, and J.-O. Chae, Efficiency of  $NO_x$  reduction in pulse-streamer discharge, in: *Proc. Int. School-Seminar Nonequilibrium Processes and Their Applications*, 8–13 September 1994, Contributer Paper, Minsk (1994), p. 103.
- 9. B. Eliasson and U. Kogelschatz, Modelling and applications of silent discharge plasmas, *IEEE Trans. Plasma Sci.*, **19**, April, No. 2, 309–323 (1991).
- A. Mizuno, Y. Yamazaki, S. Obama, and K. Okazaki, Effect of voltage waveform on partial discharge in ferroelectric pellet layer for gas cleaning, *IEEE Trans. Ind. Appl.*, 29, March/April, No. 2, 262–267 (1993).
- K. Jogan, A. Mizuno, T. Yamamoto, and J.-S. Chang, The effect of residence time on the CO<sub>2</sub> reduction from combustion flue gases by an ac ferroelectric packed bed reactor, *IEEE Trans. Ind. Appl.*, 29, September/October, No. 5, 876–881 (1993).