DECOMPOSITION OF DICHLOROETHANE VAPOR IN BARRIERLESS DISCHARGE

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Decomposition of dichloroethane vapor in the low-temperature plasma of barrierless gas discharge was investigated. High-voltage pulsed power supply forms high-voltage pulses of microsecond duration (1...2,2 ms) with a pulse repetition rate of 1...15 kHz and amplitude up to 15 kV. That voltage pulse shape was not changed by dichlorethane vapors in the air. At the same time, as dichlorethane vapor concentration was increased, so the amplitude of current pulse was decreased. Active radicals formed as a result of decomposition are easily oxidized by atomic oxygen and ozone, which are simultaneously synthesized from the air in barrierless discharge. It was shown, that dichlorethane decomposition reached 100% at the power of 100 W.

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INTRODUCTION

Gas emissions of industrial facilities are very dangerous pollutants for environment. For example, even low concentration of specific VOCs can be accumulated and may have adverse health effects ("low doses" effect). That is why the development of new, more sophisticated, methods for air purification are an actual task [1]. The low-temperature plasma of barrierless gas discharge is the best source of charged particles and active radicals, which strongly influence decomposition, and oxidation of wide range of toxic chemicals [2]. Dichloroethane is one of the typical pollutants. It simulates the species of halocarbon, a very dangerous gas contaminant. MPC for dichloroethane in the air is 1 mg/m3.

The studying of dichloroethane decomposition in the barrierless gas discharge is the main goal of the research. It allows determining the prospects of air purification from halocarbons by plasma chemical methods.

EXPERIMENTAL SETUP

Saturated gas mixture was prepared by bubbling of the air flow through the vessel with liquid dichloroethane. The initial dichloroethane concentration (CICH2CH2Cl) in the air was 1000 mg/m³. Flow rate of gas mixture through plasma chemical reactor was 0.5 l/min.

Plasma chemical reactor (PCR) of coaxial type included central electrode with the set of star-like electrodes (100 pieces, the electrode gap of 5 mm) and the tubular electrode. Volume of PCR is 1000 cm³. Both electrodes were made of stainless steel.

High-voltage pulsed power supply forms high-voltage pulses of microsecond duration (1...2,2 ms) with a pulse repetition rate of 1...15 kHz and amplitude up to 15 kV. The rise time of the pulse is 0,5...1,5 ms. The electric signals were recorded with a Tektronix TDS-2024 digital oscilloscope, bandwidth of 100 MHz.

The basic scheme of the experimental setup is shown in Fig. 1.

The power of the discharge was determined by the average current of the source and by the voltage on the primary winding of the transformer.

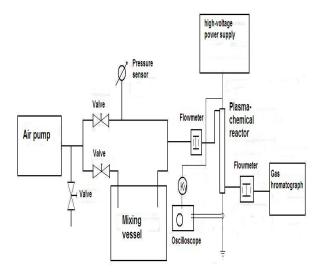


Fig. 1. Basic scheme of the experimental setup

The sample for the analysis of gas composition of plasma-chemical reaction products was taken from the air flow immediately at the outlet of plasma chemical reactor. The sample was analyzed on gas chromatograph Series II, equipped with chromatographic column Rtex-5 (30 m, 0.53mm ID, 1.5 µm df), with chemical composition of static layer of 5% diphenyl and 95% dimethyl polysiloxane, and flameionization detector of HP 19231 chromatographic column allows separating hydrocarbons of the different structure (alcohols, halocarbons, aromatic, alkanes, etc.). The flame-ionization detector allows detecting any hydrocarbon compounds with high sensitivity. Hydrogen from hydrogen generator SRI H2-40 was used as "carrier" gas. Hydrogen flow rate through the chromatographic column was 30 ml/min. The concentration of dichloroethane was determined from calculation of area under chromatographic peak. The chromatograph was calibrated using pre-prepared mixture of dichloroethane and air at well-known parameters of stationary saturation of the air with dichloroethane vapor.

RESULTS

The typical oscillogram of voltage and current pulses for discharge in the air flow saturated with dichloroethane vapors is shown in Fig. 2.

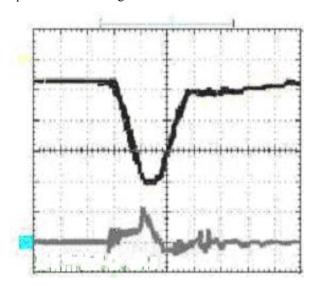


Fig. 2. Typical oscillogram (500 ns/div) of voltage (lower, 2 kV/div) and current (upper, 100 mA/div) pulses. Consumption of the mixture of dichloroethane and air vapor through the PCR is 0,5 l/min

Note, that voltage pulse shape was not changed by dichlorethane vapors in the air. At the same time, as dichlorethane vapor concentration was increased, so the amplitude of current pulse was decreased.

The amplitude of high-voltage pulse for plasmachemical reactor with the discharge gap of 5mm was 5.9 kV at the pulse-repetition frequency of 15 kHz. When amplitude of high-voltage pulse achieves 8.1 kV, the spark breakdown occurs.

As atomic oxygen and ozone are also formed in the barrierless discharge, dichlorethane decomposition products are additionally oxidized in the mixture.

$$egin{array}{c|cccc} \mathbf{H} & \mathbf{H} \\ & | & | \\ \mathbf{CI} - \mathbf{C} - \mathbf{C} - \mathbf{CI} + \mathbf{e} \Longrightarrow \mathbf{Products} \\ & | & | \\ & \mathbf{H} & \mathbf{H} \end{array}$$

As the bond, energies for dichlorethane molecule are (kJ / mol) 402.08 (C-H), 332.21 (C-Cl), 368.19 (C-C), the most probable intermediate products of dichlorethane decomposition are:

$$ClCH_2CH_2Cl + e \Rightarrow ClCH_2CH_2^* + Cl^-, \quad (1)$$

$$ClCH_2CH_2Cl + e \Rightarrow ClCH_2^* + ClCH_2^-.$$
 (2)

Active radicals formed as a result of decomposition are easily oxidized by atomic oxygen and ozone, which are simultaneously synthesized from the air in barrierless discharge.

The area of chromatographic peaks was controlled in the experiment.

The typical chromatogram of dichloroethane is shown in Fig. 3.

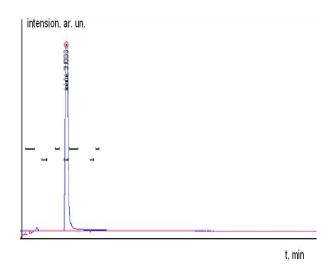


Fig. 3. Chromatogram of dichloroethane vapor

It should be noted that there are no other chromatographic peaks on chromatograms in the experiment. Only dichlorethane peak was recorded. At different power only the amplitude and the area of dichlorethane chromatographic peak were changed. The dependence of dichlorethane vapor decomposition level on the power at the air flow rate of 0,5 l/min is shown in Fig.4.

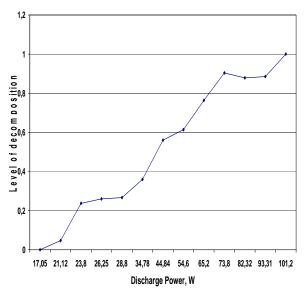


Fig. 4. Dependence of dichlorethane decomposition level on the power

While the discharge power is increased, the level of dichloroethane decomposition is increased too. It is shown in the plot at the Fig. 4. It can be assumed, that such behaviour is described according to/in accordance with Vasilyev-Kobozev-Eremin law. According to this law the rate of chemical reaction in discharge of this type is proportional to the discharge specific power.

There is no dichlorethane chromatographic peak at the power of 100 W in the experiment. Therefore, in prebreakdown regime with the voltage pulse amplitude of 7.9 kV the dichlorethane decomposition reached 100%. The exposition time for dichlorethane-air mixture in PCR was 12 s. The discharge area of plasma chemical reactor is not large.

Thus, the optimization of geometric parameters of plasma chemical reactor allows significant increasing of dichlorethane vapor decomposition effectiveness.

SUMMARY

- 1. Emissions of dichloroethane can be decomposed by low-temperature plasma gas discharge.
- 2. The level of decomposition can reach up to 100% when the discharge power comes up to 100 W.

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РАЗЛОЖЕНИЕ ПАРОВ ДИХЛОРЭТАНА В БЕЗБАРЬЕРНОМ РАЗРЯДЕ

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Исследовалось разложение паров дихлорэтана в низкотемпературной плазме безбарьерного газового разряда. Высоковольтный источник питания позволяет формировать импульсы микросекундной длительности (1...2,2 мкс) с частотой 15 кГц и амплитудой до 15 кВ. Импульс напряжения не изменялся для смеси паров дихлорэтана с воздухом относительно чистого воздуха. В то же время при росте концентрации дихлорэтана в воздухе происходило снижение амплитуды импульса тока. Активные радикалы, которые формируются в результате разложения дихлорэтана, легко окисляются атомарным кислородом и озоном, параллельно образующемся в безбарьерном разряде из воздуха. Показано, что при вложенной мощности порядка 100 Вт степень разложения дихлорэтана достигает 100 %.

РОЗКЛАДАННЯ ПАРІВ ДІХЛОРЕТАНУ У БЕЗБАР'ЄРНОМУ РОЗРЯДІ

В.І. Голота, Л.М. Завада, О.В. Котюков, Д.В. Кудін, С.В. Родіонов, А.С. Письменецький, Ю.В. Доценко

Досліджувалося розкладання парів діхлоретану у низькотемпературній плазмі безбар'єрного газового розряду. Високовольтне джерело живлення дозволяє формувати імпульси мікросекундної довжини (1...2,2 мкс) з частотою 15 кГц та амплітудою до 15 кВ. Імпульс напруги не змінюється для суміши парів діхлоретана з повітрям відносно чистого повітря. В той же час при зростанні концентрації діхлоретана в повітрі відбувалося зниження амплітуди імпульса струму. Активні радикали, що формуються внаслідок розкладання діхлоретану, легко окислюются атомарним киснем та озоном, що паралельно синтезуются з повітря. Показано, що при вкладеній потужності порядку 100 Вт ступінь розкладання діхлоретану сягає 100%.