PLASMA-BEAM DISCHARGE, DISCHARGE AND PLASMACHEMISTRY

OZONE DECAY IN CHEMICAL REACTOR WITH THE DEVELOPED INNER SURFACE: AIR-ETHYLENE MIXTURE

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The ozone decay in air-ethylene mixture was studied in a free space as well as in a chemical reactor with a developed inner surface on which ozone dissociates or absorbs. It is shown that both in the case of ozone decay in free space and in the case of ozone decay in a container with a developed inner surface, there is a range of parameters (initial concentrations of ozone and ethylene, reactor inner surface area) for which the ozone concentration behaves in time as exp(-ωt), i.e. the (pseudo) first-order kinetics of ozone decay take place.
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INTRODUCTION

As an environmentally friendly oxidant, ozone can be used in a wide range of technologies in various fields of human activity, such as agriculture [1], food production and storage [2], waste recycling [3 - 5] and other. The three major problems should be solved for each ozone based technology: the energy efficient ozone production, the ozone delivery (with minimal loss) to the interaction point, and the chemical interaction of ozone with the substance to be treated. The first problem can be solved with the help of barrierless ozonizers [6] based on the streamer discharge [7 - 10]. The paper is devoted to the solution of the second and third problem in food storage.

Fresh fruits and vegetables emit ethylene – one of plant hormones, which promotes the aging of these fresh agricultural products. Therefore, ethylene is an undesirable compound when storing agricultural produce. The traditional method for fresh-keeping of fruits and vegetables is refrigeration. However, even in refrigeration environment, ethylene is released by agricultural products themselves or other sources. The residual ethylene can accelerate maturation and corruption of agricultural products and thus it should be wiped away. So, the ethylene concentration reducing in the containers for storage and transportation of perishable products is important task for keeping freshness of fruits and vegetables.

Conventional techniques for ethylene removal include adsorption, K_2MnO_4 oxidation, catalytic oxidation, photo-catalysis and some biological methods. These methods have unavoidable drawbacks: adsorbent may be damaged because of aerosols, K_2MnO_4 oxidation and catalytic oxidation are complicated, photo-catalysis and biological method have low removal rates. An alternative approach to keep fruits and vegetables fresh is usage of nonthermal plasma [11 - 14]. The dielectric barrier discharge, corona discharge, glow discharge or, in our case, barrierless streamer discharge can be applied to decompose ethylene and other volatile organic compounds. This approach has many advantages: moderate operation conditions (normal temperature and atmospheric pressure), low cost and plasma reactors compactness. As all these discharges work with

air-ethylene mixture, the ozone is generated. Ozone can be feeded into the container for the transportation of perishable products and can serve as an additional reactant for ethylene decomposition. Ozone decay and ethylene decomposition in the free space and in the container for the transportation of fruits and vegetables were investigated in this paper. The loaded container can be considered as a chemical reactor with a developed inner surface on which the ozone dissociates or absorbs. It was found that both in the case of ozone decay in free space and in the case of ozone decay in a container with a developed inner surface, there is a range of parameters (initial concentrations of ozone and ethylene, gas temperature, reactor inner surface area) for which the ozone concentration behaves in time as exp(-δt), i.e. the (pseudo) first-order kinetics of ozone decay take place.

OZONE DECAY IN AIR-ETHYLENE MIXTURE

The ethylene ozonolysis is proceed via a primary ozonide (Fig. 1), which then decomposes to give a carbonyl product together with a Griegee intermediate [15]. The Criegee intermediate may decay by one of following ways [16]: it may be stabilised by a collision with a third body to give a carbonyl oxide (CH_2O_2), it may decompose to give OH radicals, and it may react with the carbonyl product to give a secondary ozonide, which is unlikely. The secondary ozonide and carbonyl oxide will react further to yield stable products.
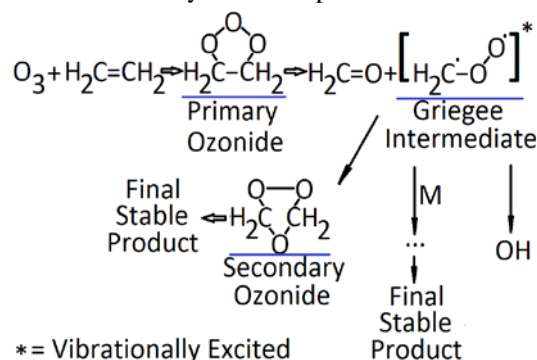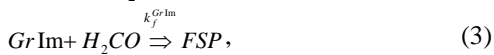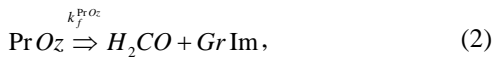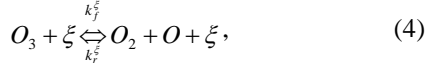

Fig. 1. The ozonolysis of ethylene

The ozonolysis of ethylene can be summarized as:

$$C_2H_4 + O_3 \xrightarrow{k_f^{Al}} \Pr Oz, \qquad (1)$$

$$\Pr Oz \xrightarrow{k_f^{\Pr Oz}} H_2CO + Gr\,Im, \qquad (2)$$

$$Gr\,Im + H_2CO \xrightarrow{k_f^{Gr\,Im}} FSP, \qquad (3)$$

where $\Pr Oz = C_2H_4O_3$ is the primary ozonide, $Gr\,Im = H_2CO_2$ is the Griegee intermediate, and FSP are the final stable products.

The conventional scheme of ozone decay in air [2 - 4, 17] can be modified to include the ozonolysis of ethylene according to (1) - (3):

$$O_3 + \xi \underset{k_r^{\xi}}{\overset{k_f^{\xi}}{\Longleftrightarrow}} O_2 + O + \xi, \qquad (4)$$

$$O_3 + O \xrightarrow{k_f^{o}} 2O_2, \qquad (5)$$

$$O_3 + Al \xrightarrow{k_f^{*Al}} FSP. \qquad (6)$$

In the equation (4) $\xi = \{N_2, O_2, H_2O, O_3, Al=C_2H_4, CO_2, He, Ar, N_2O\}$. The abbreviation «Al» means alkenes – C_nH_{2n}, in our case $Al = C_2H_4$. $k_f^{\xi}(T)$ is the rate constant of the forward reaction. It depends on the temperature T like the rest of the reaction rate constants. $k_r^{\xi}(T)$ is the reverse reaction rate constant. In the equations (5), (6) $k_f^{o}(T)$ and $k_f^{*Al}(T)$ are the forward reaction rate constant.

The forward reaction in (4) shows unimolecular ozone decay. This reaction is not elementary. It consists of a multi-stage process which includes activation and decay of the excited molecule through the activated complex. The reverse reaction is also not elementary. It flows in two biomolecular stages: formation of excited ozone with subsequent relaxation. The reaction in (5) is exothermic. The excess energy is distributed over the vibrational degrees of freedom of the oxygen molecule. As a rule, vibrationally excited oxygen relaxes to the ground state.

The system of kinetic equations for (4) – (6), if $\xi = \{N_2, O_2, H_2O, O_3, Al=C_2H_4\}$, generally is the following:

$$\frac{dC_{O_3}}{dt} = -C_{O_3} \sum_{\xi} k_f^{\xi} C_{\xi} + C_{O_2} C_O \sum_{\xi} k_r^{\xi} C_{\xi} - k_f^{o} C_{O_3} C_O - k_f^{*Al} C_{O_3} C_{Al},$$
$$(7)$$

$$\frac{dC_{O_2}}{dt} = C_{O_3} \sum_{\xi} k_f^{\xi} C_{\xi} - C_{O_2} C_O \sum_{\xi} k_r^{\xi} C_{\xi} + 2k_f^{o} C_{O_3} C_O, \quad (8)$$

$$\frac{dC_O}{dt} = C_{O_3} \sum_{\xi} k_f^{\xi} C_{\xi} - C_{O_2} C_O \sum_{\xi} k_r^{\xi} C_{\xi} - k_f^{o} C_{O_3} C_O, \qquad (9)$$

$$\frac{dC_{Al}}{dt} = -k_f^{*Al} C_{O_3} C_{Al}, \qquad (10)$$

where $C_{\xi}(t)$ is a concentration of ξ. For $\xi = \{N_2, H_2O\}$, the density of particles $C_{\xi}(t)$ may depend on time only through the initial conditions. Therefore, $C_{N_2} = $ const, $C_{H_2O} = $ const. The following equation can be obtained using the method of steady-state concentrations for O:

$$C_o(t) = \frac{C_{O_3}(t) \sum_{\xi} k_f^{\xi} C_{\xi}(t)}{C_{O_2}(t) \sum_{\xi} k_r^{\xi} C_{\xi}(t) + k_f^{o} C_{O_3}(t)}. \qquad (11)$$

It is convenient to introduce the following notations:

$$F(t,T) = \sum_{\xi} k_f^{\xi}(T) C_{\xi}(t), \; G(t,T) = \sum_{\xi} k_r^{\xi}(T) C_{\xi}(t). \quad (12)$$

The equations (7), (8) may be presented in a simple form:

$$\frac{dC_{O_3}}{dt} = -k_f^{*Al} C_{O_3} C_{Al} - 2\frac{k_f^{o} F}{GC_{O_2} + k_f^{o} C_{O_3}} C_{O_3}^2, \qquad (13)$$

$$\frac{dC_{O_2}}{dt} = 3\frac{k_f^{o} F}{GC_{O_2} + k_f^{o} C_{O_3}} C_{O_3}^2. \qquad (14)$$

The numerical estimations of F and G, using the rate constants of the corresponding reactions from [17], for the atmospheric pressure and the temperature $T \sim 300$ K, for the mass ozone concentration $MC_{O_3} < 20$ g/m^3 and water concentration $MC_{H_2O} < 25$ g/m^3, show that the main input to F will be achieved due to ozone decay at the collision with nitrogen. Ozone collision with oxygen and water gives the input to F by several times lower. Ozone collisions with ozone and ethylene give the input by two orders lower than the ozone decay on nitrogen. This allows highly accurate calculation of F in accordance with the initial densities of nitrogen, water and oxygen – $F \approx k_f^{N_2}(T) \cdot C_{N_2} + k_f^{H_2O}(T) \cdot C_{H_2O} + k_f^{O_2}(T) \cdot C_{O_2}$. A similar analysis can be carried out for G. As a result, G with high accuracy, as well as F, does not depend on time and is defined by the initial bulk densities of nitrogen, water and oxygen – $G \approx k_r^{N_2}(T) \cdot C_{N_2} + k_r^{H_2O}(T) \cdot C_{H_2O} + k_r^{O_2}(T) \cdot C_{O_2}$.

The equations (13), (14) and (10) show that in general case the ozone decay is described by the variable order kinetics, from the 1-st to the 2-nd order depending on the parameters of the problem. If $k_f^{o} C_{O_3} \gg GC_{O_2}$ (low pressure), ozone decay is described by the first order kinetics: $dC_{O_3}/dt = -\alpha C_{O_3} C_{Al} - \beta C_{O_3}$, where $\alpha = -k_f^{*Al} = $ const, $\beta = 2k_f^{o} F / k_f^{o} \approx $ const. In the opposite case, if $k_f^{o} C_{O_3} \ll GC_{O_2}$ (atmospheric pressure), ozone decay is described by the second order kinetics: $dC_{O_3}/dt = -\alpha C_{O_3} C_{Al} - \delta C_{O_3}^2$, where $\delta = 2k_f^{o} F / GC_{O_2} \approx $ const. The latter case is the most interesting; this is the ozone decay and the ethylene ozonolysis at atmospheric pressure:

$$\frac{dC_{O_3}}{dt} = -\alpha C_{O_3} C_{Al} - \delta C_{O_3}^2, \; \frac{dC_{Al}}{dt} = -\alpha C_{O_3} C_{Al}. \quad (15)$$

To analyze equations (15) from a bird's eye view, these equations were solved numerically for the initial conditions $C_{O_3}(0) \ll C_{Al}(0)$ (Fig. 2) and $C_{O_3}(0) \gg C_{Al}(0)$ (Fig. 3) and different ratios of the coefficients α and δ: $\alpha \sim \delta$, $\alpha \ll \delta$, and $\alpha \gg \delta$. As can be seen from Fig. 2, for $C_{O_3}(0) \ll C_{Al}(0)$, ozone decays as exp(-ωt). However, ozone is not sufficient to substantially reduce the ethylene concentration. It was this case that was studied experimentally in [18]. In the opposite case (see Fig. 3), when $C_{O_3}(0) \gg C_{Al}(0)$, the ethylene concentration can be substantially reduced due to the ethylene ozonolysis.

The particles decay on the surface can be included in the equations (7) - (10). To do this, the continuity equations should be integrated over the volume in the same way as was done in [2], [4].

First of all, let us take into account the decay on the wall for atomic oxygen:

$$\frac{dC_O}{dt} = FC_{O_3} - GC_{O_2}C_O - k_f^o C_{O_3} C_O - \beta_O C_O, \quad (16)$$

where $\beta_O = \alpha v_O \gamma_O S/V$ is the rate constant of the atomic oxygen absorption on the surface, α is the coefficient considering the problem geometry, v_O is the particle (thermal) velocity, γ_O is the probability of the particle loss on the surface, V is the vessel volume, and S is an inner surface.
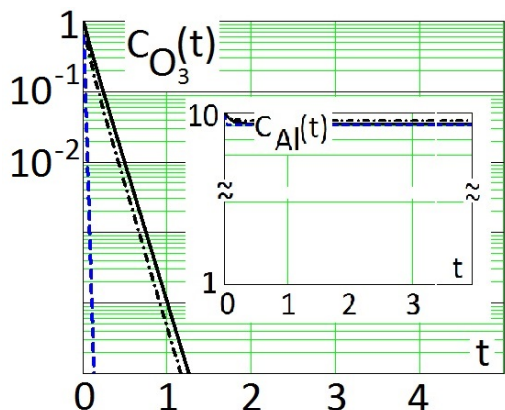

Fig. 2. Ozone decay and ethylene ozonolysis at atmospheric pressure. $C_{O_3}(t)$ – ozone concentration vs time, $C_{Al}(t)$ – ethylene concentration vs time. Initial conditions: $C_{O_3}(0) \ll C_{Al}(0)$. Solid black line – $\alpha \sim \delta$, dot-dashed black line – $\alpha \ll \delta$, dashed blue line $\alpha \gg \delta$
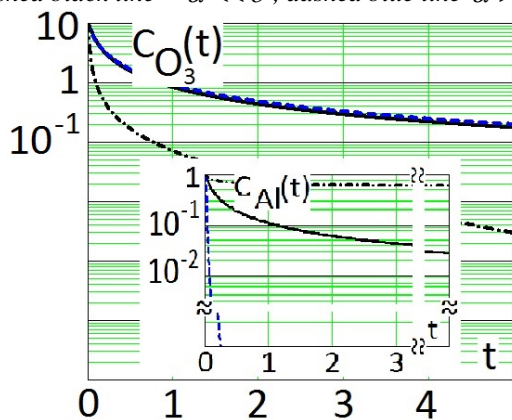

Fig. 3. Ozone decay and ethylene ozonolysis at atmospheric pressure. $C_{O_3}(t)$ – ozone concentration vs time, $C_{Al}(t)$ – ethylene concentration vs time. Initial conditions: $C_{O_3}(0) \gg C_{Al}(0)$. Solid black line – $\alpha \sim \delta$, dot-dashed black line – $\alpha \ll \delta$, dashed blue line $\alpha \gg \delta$

The following equation can be obtained using the method of stationary concentrations for O:

$$C_O(t) = \frac{FC_{O_3}}{GC_{O_2} + k_f^o C_{O_3} + \beta_O}. \quad (17)$$

The following equations for concentrations of O₃, O₂ and C₂H₄ can be obtained from equations (7), (8), (10), taking into account (17):

$$\frac{dC_{O_3}}{dt} = -\beta_{O_3}C_{O_3} - k_f^{*Al}C_{O_3}C_{Al} - \frac{2k_f^o C_{O_3} + \beta_O}{GC_{O_2} + k_f^o C_{O_3} + \beta_O}FC_{O_3}, \quad (18)$$

$$\frac{dC_{O_2}}{dt} = -\beta_{O_2}C_{O_2} + \frac{3k_f^o C_{O_3} + \beta_O}{GC_{O_2} + k_f^o C_{O_3} + \beta_O}FC_{O_3}, \quad (19)$$

$$\frac{dC_{Al}}{dt} = -\beta_{Al}C_{Al} - k_f^{*Al}C_{O_3}C_{Al}, \quad (20)$$

where $\beta_{O_3} = \alpha v_{O_3}\gamma_{O_3}S/V$, $\beta_{O_2} = \alpha v_{O_2}\gamma_{O_2}S/V$, and $\beta_{Al} = \alpha v_{Al}\gamma_{Al}S/V$ are the rate constants of the particles losses on the surface for ozone, oxygen and ethylene, respectively. Let us consider the limiting cases.

If $\beta_O \gg 2k_f^o C_{O_3}$, and $\beta_O \gg GC_{O_2} + k_f^o C_{O_3}$, the equation (18) gives:

$$\frac{dC_{O_3}}{dt} = -\beta_{O_3}C_{O_3} - k_f^{*Al}C_{O_3}C_{Al} - FC_{O_3}. \quad (21)$$

If $\beta_O \ll 2k_f^o C_{O_3}$, and $\beta_O \ll GC_{O_2} + k_f^o C_{O_3}$, the equation (18) gives:

$$\frac{dC_{O_3}}{dt} = -\beta_{O_3}C_{O_3} - k_f^{*Al}C_{O_3}C_{Al} - \frac{2k_f^o FC_{O_3}^2}{GC_{O_2} + k_f^o C_{O_3}}. \quad (22)$$

If $GC_{O_2} \gg k_f^o C_{O_3}$, equation (22) gives:

$$\frac{dC_{O_3}}{dt} = -\beta_{O_3}C_{O_3} - k_f^{*Al}C_{O_3}C_{Al} - \frac{2k_f^o FC_{O_3}^2}{GC_{O_2}}. \quad (23)$$

If $GC_{O_2} \ll k_f^o C_{O_3}$, equation (22) gives:

$$\frac{dC_{O_3}}{dt} = -\beta_{O_3}C_{O_3} - k_f^{*Al}C_{O_3}C_{Al} - 2FC_{O_3}. \quad (24)$$

If $\beta_O \gg 2k_f^o C_{O_3}$, and $\beta_O \ll GC_{O_2} + k_f^o C_{O_3}$, the equation (18) gives:

$$\frac{dC_{O_3}}{dt} = -\beta_{O_3}C_{O_3} - k_f^{*Al}C_{O_3}C_{Al} - \frac{\beta_O FC_{O_3}}{GC_{O_2} + k_f^o C_{O_3}}. \quad (25)$$

If $GC_{O_2} \gg k_f^o C_{O_3}$, equation (25) gives:

$$\frac{dC_{O_3}}{dt} = -\beta_{O_3}C_{O_3} - k_f^{*Al}C_{O_3}C_{Al} - \frac{\beta_O FC_{O_3}}{GC_{O_2}}. \quad (26)$$

If $GC_{O_2} \ll k_f^o C_{O_3}$, equation (25) gives:

$$\frac{dC_{O_3}}{dt} = -\beta_{O_3}C_{O_3} - k_f^{*Al}C_{O_3}C_{Al} - \frac{\beta_O F}{k_f^o}. \quad (27)$$

If $\beta_O \ll 2k_f^o C_{O_3}$, and $\beta_O \gg GC_{O_2} + k_f^o C_{O_3}$, the equation (18) gives:

$$\frac{dC_{O_3}}{dt} = -\beta_{O_3}C_{O_3} - k_f^{*Al}C_{O_3}C_{Al} - \frac{2k_f^o FC_{O_3}^2}{\beta_O}. \quad (28)$$

Thus, the ozone decomposition in an air-ethylene mixture is described by three types of equations:

$$\frac{dC_{O_3}}{dt} = -\alpha C_{O_3}C_{Al} - \beta C_{O_3} - \delta C_{O_3}^2, \quad (29)$$

$$\frac{dC_{O_3}}{dt} = -\alpha C_{O_3}C_{Al} - \beta C_{O_3} - \delta C_{O_3}, \quad (30)$$

$$\frac{dC_{O_3}}{dt} = -\alpha C_{O_3}C_{Al} - \beta C_{O_3} - \delta. \quad (31)$$

The equation for ethylene ozonolysis taking into account its absorption on the wall has the form:

$$\frac{dC_{Al}}{dt} = -\alpha C_{O_3}C_{Al} - \mu C_{Al}. \quad (32)$$

The most interesting cases are (29), (30) and (32). These are the ozone decay and the ethylene ozonolysis at atmospheric pressure. To analyze equations (29), (32) from a bird's eye view, these equations were solved numerically for the initial conditions $C_{O_3}(0) << C_{Al}(0)$ (see Fig. 2) and $C_{O_3}(0) >> C_{Al}(0)$ (see Fig. 3) and different ratios of the coefficients α, β, δ, and μ: $\alpha \sim \beta \sim \delta \sim \mu$, $\alpha \sim \beta \sim \delta >> \mu$, $\alpha \sim \beta \sim \delta << \mu$, $\alpha \sim \beta >> \delta \sim \mu$, etc. As can be seen from Fig. 4, for $C_{O_3}(0) << C_{Al}(0)$, there is a range of parameters where ozone and ethylene decay as exp(-ωt). Same in the opposite case (Fig. 5), when $C_{O_3}(0) >> C_{Al}(0)$. In both cases, the ethylene concentration decreases with time due to decay on the inner surface and ozonolysis.
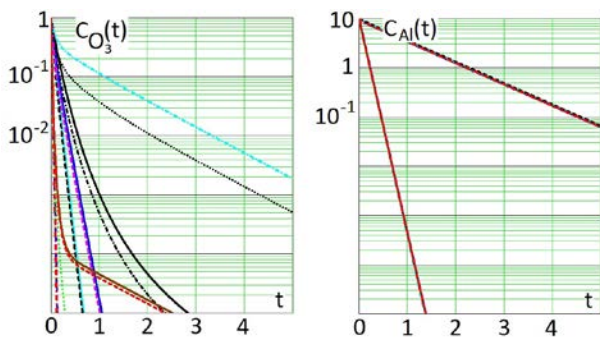

Fig. 4. Ozone decay and ethylene ozonolysis at atmospheric pressure. $C_{O_3}(t)$ − ozone concentration vs time,

$C_{Al}(t)$ − *ethylene concentration vs time.*

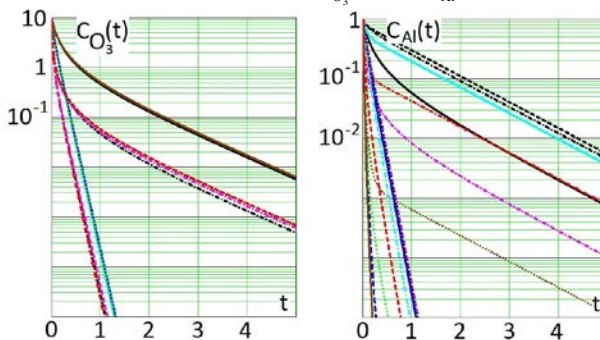*Initial conditions:* $C_{O_3}(0) << C_{Al}(0)$

Fig. 5. Ozone decay and ethylene ozonolysis at atmospheric pressure. $C_{O_3}(t)$ − ozone concentration vs time,

$C_{Al}(t)$ − *ethylene concentration vs time.*

Initial conditions: $C_{O_3}(0) >> C_{Al}(0)$

Analysis (30), (31) gives similar results: there is a range of parameters for which ozone decays as exp(-ωt).

CONCLUSIONS

The ozone decay in air-ethylene mixture was studied in a free space as well as in a chemical reactor with a developed inner surface on which ozone dissociates or absorbs. The kinetic equations of ozone decay in the ozone-ethylene mixture at atmospheric pressure were obtained and analyzed. It is shown that, in the general case, the ozone decay kinetics is of the order not higher than the second. However, under certain conditions (the developed inner reactor surface, high initial ethylene

concentrations), the second-order kinetics degenerates into first-order kinetics. So, it is shown that both in the case of ozone decay in free space and in the case of ozone decay in a container with a developed inner surface, there is a range of parameters (initial concentrations of ozone and ethylene, reactor inner surface area) for which the ozone concentration behaves in time as exp(-ωt), i.e. the (pseudo) first-order kinetics of ozone decay take place.

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РАСПАД ОЗОНА В ХИМИЧЕСКОМ РЕАКТОРЕ С РАЗВИТОЙ ВНУТРЕННЕЙ ПОВЕРХНОСТЬЮ: ВОЗДУШНО-ЭТИЛЕНОВАЯ СМЕСЬ

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Исследована кинетика распада озона в воздушно-этиленовой смеси в неограниченном пространстве и в химическом реакторе с развитой внутренней поверхностью, на которой озон может распадаться. Показано, что как в свободном неограниченном пространстве, так и в случае распада озона в контейнере с развитой внутренней поверхностью, существует область параметров (начальные концентрации озона и этилена, площадь внутренней поверхности реактора), для которых концентрация озона ведет себя во времени как exp(-ωt), что соответствует кинетике (псевдо) первого порядка.

РОЗПАД ОЗОНУ В ХІМІЧНОМУ РЕАКТОРІ З РОЗВИНЕНОЮ ВНУТРІШНЬОЮ ПОВЕРХНЕЮ: ПОВІТРЯНО-ЕТИЛЕНОВА СУМІШ

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Досліджено кінетику розпаду озону в повітряно-етиленовій суміші в необмеженому просторі та у хімічному реакторі з розвиненою внутрішньою поверхнею, на якій озон може розпадатися. Показано, що як у вільному необмеженому просторі, так і в разі розпаду озону в контейнері з розвиненою внутрішньою поверхнею, існує область параметрів (початкові концентрації озону і етилену, площа внутрішньої поверхні реактора), для яких концентрація озону поводить себе у часі як exp(-ωt), що відповідає кінетиці (псевдо) першого порядку.