

Destructive oxidation of ethanol in the corona discharge reactor

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The results of investigation of ethanol destructive oxidation (model aliphatic alcohol) in a corona discharge reactor are presented. The process was performed at the temperature of 303 K in the corona discharge generator – the reactor system manufactured in our laboratory. The process temperature was kept constant by cooling down the reactor with a stream of air. The measurements were carried out using the following process parameters: the inlet ethanol concentration in the stream of gases in the range of 0.0028 to 0.132 mol/m³ (0.128 ÷ 6.086 g/m³), the gas flow velocity in the range of 0.15 – 0.33 m³/h (space velocity in the range of 1220 ÷ 2680 m³/(m³_R·h)) and the power supply to the reactor ranged from 1.6 to 86.4 W. The active volume of the reactor was 1.23·10⁻⁴ m³. The phenomenological method was applied for the description of the process. It was based on the assumptions that the reaction rate can be described by the first order equation in relation to the ethanol concentration and the design equation of flow tubular reactor can be applied for the description of corona reactor. The usefulness of this model was estimated using statistical methods for the analysis of the experimental results. The Statistica 6.0 software was used for this application. The first stage of this analysis showed the dependencies between the considered variables, whereas the second stage was to find the equations describing the influence of the selected process parameters on the rate of ethanol destruction. The parameters of A and B of apparent constant rate equation given in the form of $Z = A \cdot \exp(-B/P)$ were also determined.

The results of the investigations indicated that the applied corona discharge generator – reactor system assures a high efficiency of purification of the air and industrial waste gases contaminated by ethanol. The ethanol destruction degree of $\alpha_r = 0.9$ was obtained at the power supply to the reactor amounting to 650 kW/m³_R per unit of its active volume. The final products of the reaction were only the harmless carbon dioxide and water vapour. It has been stated that the rate of the destructive oxidation of ethanol reaction is well described by the first order equation in relation to the ethanol concentration. Under isothermal conditions, the reaction rate also depends on the power supply to the reactor. This dependence is well described by the empirical equation $Z = 3,233 \cdot \exp(-82,598/P)$.

The obtained results also indicated that the method of destructive oxidation of ethanol in the corona discharge reactor can be useful for the removal of ethanol and probably other aliphatic alcohols from different gases. The described method of calculation of the real rate of the process can be successfully used in the design of corona discharge reactors applied for such processes.

Keywords: corona discharge reactor, destructive oxidation of ethanol, kinetics of reaction.

INTRODUCTION

The volatile organic compounds (VOCs), including aliphatic alcohols, are emitted from various industrial, commercial and domestic sources. They are considered as the major contributors to air pollution and photochemical formation of smog¹. Ethanol is one of the dominating VOCs in some segments of the printing industry, where it is used as a component of solvents and paints, in distilling industry and in exhaust gases from ethanol-fuelled vehicles^{1,2}. Although the use of ethanol as fuel can reduce the emission of conventional pollutants such as CO, it can also increase the direct emission of unburnt ethanol³.

The destructive oxidation of the volatile organic compounds in the air or effluent gases using the corona discharge is an effective method of air and industrial waste gases purification, which has been developed in recent years. This process is carried out in the corona reactor, where the pollutants are exposed to the action of low-temperature plasma and ozone, which are formed in the reactor as the result of corona discharge. Under such

conditions, most often the organic compounds are completely oxidized. When the pollutants contain in their structures only C, H and O, the harmless water vapour and carbon dioxide are formed as the reaction products. This method is particularly useful when the source of pollutants emission works periodically or at irregular intervals. Moreover, it is cheaper than other methods because the process proceeds effectively at low temperatures in the range of 303 – 353 K^{4-8,12,14,17}.

In the literature, there are few data regarding the destructive oxidation of aliphatic alcohols in the corona discharge reactors. In paper⁴ the results of preliminary investigations of the destruction of isopropyl alcohol in the corona discharge generator- reactor system manufactured in the our laboratory were presented. The obtained results were taken as the basis for further investigations of the destructive oxidation of isopropyl alcohol⁵ and n-butyl acetate^{6,7}. In these papers the applied systems of apparatus, different constructions of the corona discharge reactor and especially the electrodes' systems for reactors, were described. The destructive oxidation of isopropyl

alcohol in the corona reactor was also presented in paper⁹. When the concentration of alcohol in the stream of gases was 100 ppm and the residence time in the reactor was in the range of 0.20 – 0.75 s, the high degree of destruction of 0.995 was obtained. However at higher ethanol concentration (1000 ppm), the conversion degree was 0.65 for the residence time of 0.25 s and 0.99 for residence time of 1.5 s. The power consumption by the corona reactor was 16 W, and it was kept constant for both measurements.

In the paper¹⁰ the methanol decomposition process in the corona reactor was investigated. It was stated that methanol below 50 ppm in air stream containing 2 mg/dm³ of water vapour was decomposed completely by using dielectric barrier discharge (DBD) under discharge conditions as $U = 16$ kV and $f = 100$ Hz. The results of the numerical analysis indicated that the most important radical to decompose the methanol was $\cdot\text{OH}$. Sobacchi et al.¹¹ present the results of the study on the removal of VOC mixtures typically present in the exhaust streams of wood processing industry. Methanol, acetone, dimethyl sulphide and α – pinene were selected as representative VOC compounds. The measurements were carried out using the streamer corona discharge reactor for individual VOCs and their mixtures, varying input concentration (5 to 1000 ppm), temperature (298 to 473 K), and relative humidity (0 to 100%). The gas flow rate was kept at 2 dm³/min resulting in the residence time about 13 s. It was found that the destruction and removal efficiencies (DREs) above 98% were obtained for all the studied compounds and they depended on the temperature, relative humidity and the concentration of VOC. The energy costs of VOC removal of the compound increased in the order: dimethyl sulphide, methanol, α – pinene, acetone. It was found that an increase of the temperature and moderate humidity resulted in the reduction of decomposition costs. The results of chromatographic analysis of the outlet gases from the reactor showed that no organic byproducts were produced during the destruction of methanol, whereas acetone and dimethyl sulphide decomposition resulted in the formation of a small amount of methanol. In the case of α -pinene destruction the small amounts of methanol and significantly larger quantities of acetone were produced. The acetone concentration depends both on the initial concentration of α – pinene and power supply to the reactor.

The applications of dielectric barrier discharges for the removal of other VOCs from the exhaust gases and air were presented in papers^{12–23}. The different constructions of corona reactors^{12–17} and corona reactors containing a heterogeneous catalyst in or near the discharge zone^{18–23} were applied. It was reported^{18, 20–22} that the introduction of the catalyst enhanced the conversion of pollutants and allowed to obtain the higher CO₂ selectivity.

The aim of this study was to determine the efficiency of the corona discharge reactor manufactured in the laboratory, in the process of the purification of the air contaminated by ethanol, taken as a representative type of the aliphatic alcohols. Moreover, the macrokinetics of the destructive oxidation process of ethanol in the corona discharge reactor was also described.

THEORETICAL BASIS

The mathematical model presented in details in work⁴ was applied to describe the macrokinetics of the investigated process. The assumptions of this model were as follows:

– the operation of the corona discharge reactor can be described by the design equation of the flow tubular reactor²⁴:

$$\dot{F}_{0v} \cdot c_{0i} \cdot d\alpha_i = r_i \cdot dV_R \quad (1)$$

– the reaction rate in the corona discharge reactor, under isothermal conditions, only depends on the power supply to the reactor and the conversion degree and formally it can be written as

$$r_i = f(P, \alpha_i) = \varphi(P) \cdot \psi(\alpha_i) \quad (2)$$

– the reaction rate is described by the first order equation in relation to the concentration of the pollutant and the process takes place at a constant temperature, thus the constituent functions of equation (2) can be written as follows:

$$\psi(\alpha_i) = c_{0i} \cdot (1 - \alpha_i) \quad (3)$$

$$\varphi(P) = Z = A \cdot \exp\left(-\frac{B}{P}\right) \quad (4)$$

and after substitution these functions into formula (2) it has the form:

$$r_i = A \cdot \exp\left(-\frac{B}{P}\right) \cdot c_{0i} \cdot (1 - \alpha_i) = Z \cdot c_{0i} \cdot (1 - \alpha_i) \quad (5)$$

– the function $Z = \varphi(P)$ was treated, like in the classic kinetics, as the apparent rate constant of reaction at the known, constant temperature; the parameters of this equation are determined on the basis of the experimental results using the dependence $\ln Z = f(1/P)$.

By substitution of equation (5) into formula (1) and followed by its integration and appropriate transformations allows to obtain the design equation of the corona discharge reactor in the form:

$$\alpha_i = 1 - \exp\left(-\frac{Z}{\dot{F}_{0v}} \cdot V_R\right) \quad (6)$$

The value of the Z function, for the applied power supply to the reactor and the constant reaction temperature, can be calculated from formula (6), after its transformation into the form:

$$Z = \varphi(P) = \frac{1}{\tau_z'} \cdot \ln\left(\frac{1}{1 - \alpha_i}\right) \quad (7)$$

where $\tau_z' = V_R / \dot{F}_{0v}$ is the total residence time of gases inside the reactor.

EXPERIMENTAL

The investigations were carried out using the experimental set-up schematically shown in Figure 1.

The feed air from the pump (1) was divided into two streams. One stream was bubbled through ethanol in a washer (2). The volume flow of this stream was controlled by a microvalve. The temperature of the washer was held constant using a water bath. The air was saturated with

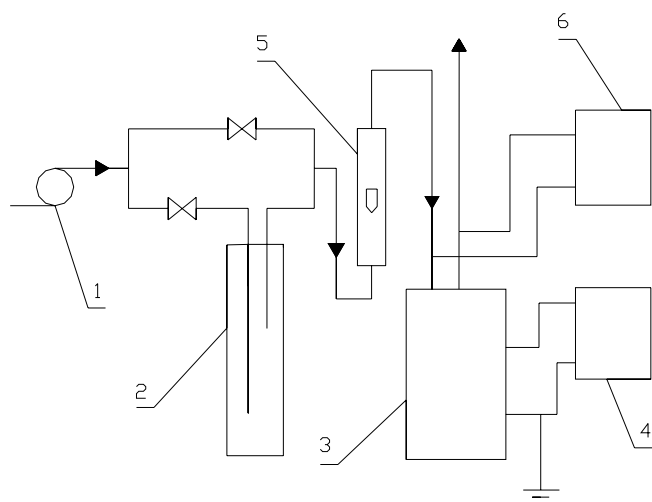


Figure 1. The experimental set-up: 1 – pump, 2 – washer with ethanol, 3 – corona discharge reactor, 4 – corona discharge generator, 5 – flowmeter, 6 – gas chromatograph

ethanol vapour in the washer. The second stream, controlled by another microvalve, was pure air. Both streams were mixed. By using the microvalves system, the proper contribution of the feed air and the required concentration of ethanol in the air-ethanol mixture were obtained. The total volume flow rate of the mixture was measured by a flowmeter (5) and next the mixture was then introduced into the reactor (3), which was connected to a corona discharge generator (4). The reactor was cooled down with a stream of air from a small fan. The reaction temperature was controlled using three thermoelements placed on the external surface of the reactor. It was stated, that the temperature was constant (303 ± 1 K) during each measurement. It was assumed, that the reactor operates under isothermal conditions. The sampling ports were fitted at the inlet and the outlet of the reactor. The samples of gases for analysis were taken from these ports, using a syringe.

The inlet and outlet concentrations of ethanol in the stream of gases were determined using a gas chromatograph CHROM-5 equipped with FID detector and three-meter column packed with Porapak QS. The column temperature reached 383 K. The chromatograph was connected to a computer which recorded the chromatograms and calculated the surface areas of the peaks in the chromatograms.

With the exception of unreacted ethanol, no other possible organic by-products of the ethanol oxidation such as acetone, acetaldehyde, acetic acid, diethylether or ethylene were found in the outlet gases. Only CO_2 and H_2O were the products of the reaction.

The operation parameters of the corona discharge generator, applied in the study, were as follows: maximum output high-voltage of 15 kV, maximum output power of 500 W, polarization high-voltage period of 150 μs , duration of corona impulse of 2.5 μs .

The reactor consisted of two pipes concentrically packed in the teflon heads (fixing elements) equipped with connector pipes to let the gas into the reaction zone and to take its products off:

– external glass pipe 0.22 m in length, inner diameter $D = 0.0262$ m and wall thickness = 0.002 m, inside of which an earthing electrode was placed.

– internal quartz pipe 0.22 m in length, outer diameter $d = 0.018$ m and wall thickness = 0.002 m (inner diameter $d_1 = 0.014$ m), the pipe contained high voltage electrode.

The earthing electrode was made of stainless steel wire 0.001 m thick in a form of spiral and was pulled at the full length of the reactor. The high-voltage electrode was a stainless steel wire brush 0.10 m in length and 0.016 m in diameter. It was placed in the middle of the inner pipe of the reactor.

The electrodes shaped and placed in the reactor in this way ensured the heterogeneity of the electric field, throughout the whole length of the reactor along the high-voltage electrode. The observed shape of luminescence of corona discharges depended on the applied input high-voltage. When the input voltage was low, it appeared in the shape of a ribbon along the spiral of the earthing electrode, but if the higher voltage was applied the ribbon coils aggregated. Under such conditions the lines of corona discharges were more distinctive. For both cases the total residence time in the reactor was the same. The schematic diagram of the corona discharge reactor used in the investigations is illustrated in Figure 2.

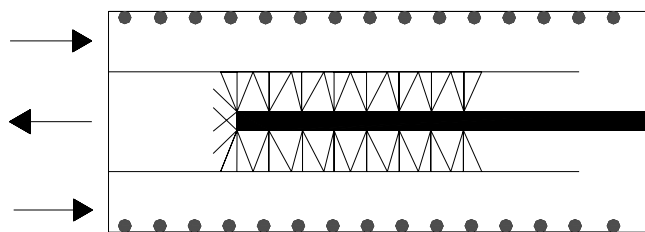


Figure 2. A scheme of the corona discharge reactor

The air-ethanol mixture was introduced into the reactor between its external and internal pipes and was then passed through the first inactive zone of the reactor to the corona discharge zone (active zone of the reactor), where the reactions of the formation of ozone and destruction of ethanol were initiated. Then, the process proceeded in the second inactive zone and in the internal pipe, where unreacted ethanol and its partially oxidized products were subjected to a further oxidation with ozone. This fact together with the effect of UV-rays formed in the reactor as the result of corona discharges, resulted in a longer residence time and higher efficiency of ethanol destruction.

To calculate the total residence time in the reactor, which contains one active and two symmetrically inactive zones and additionally one inactive zone inside the internal pipe, the following equation was used:

$$\tau'_z = \sum_{k=1}^n \tau'_{zr} + \sum_{j=1}^m \tau'_{zn} = \frac{\pi}{4 \cdot \dot{F}_{0v}} \cdot \left[(D^2 - d^2) \left(\sum_{k=1}^n L' + \sum_{j=1}^m L'' \right) + d_1^2 \cdot L \right] \quad (8)$$

The effect of gas flow rate on the residence time in the corona discharge zone of the reactor τ'_{zr} and on the total residence time in the reactor τ'_z , for the reactor $L = 0.22$ m in length and $n = 1$ (the number of active zones between the external and internal pipes of the reactor), $L' = 0.1$ m, but $m = 2$ (the number of the inactive zones between the external and internal pipes of the reactor) and

$L'' = 0.06$ m is presented in Figure 3. The active volume of the reactor was $V_R = 1.23 \cdot 10^{-4}$ m³.

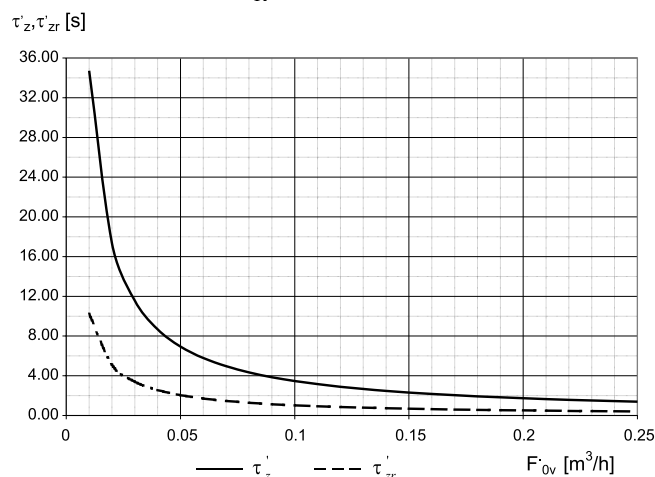


Figure 3. The residence time in the corona discharge zone of the reactor and the total residence time in the reactor versus gas flow rate

As a measure of the efficiency of destructive ethanol oxidation in the applied corona discharge generator – reactor system, the ethanol destruction degree was taken. It was expressed as:

$$\alpha_i = \frac{c_{0i} - c_i}{c_{0i}} \quad (9)$$

The power supply to the reactor was calculated using the equation:

$$P = U \cdot I \quad (10)$$

The investigations of the efficiency of the used corona discharge reactor used in the destructive oxidation of ethanol process were carried out over a wide range of the process parameters. The gas flow velocity \dot{F}_{0v} was changed in the range of $0.15 \div 0.33$ m³/h ($4.17 \cdot 10^{-5} \div 9.17 \cdot 10^{-4}$ m³/s), using the space velocity \dot{F}_{0v}' of $1.22 \cdot 10^3 \div 2.68 \cdot 10^3$ m³/(m³_R·h). The power supply to the reactor was in the range of $1.6 \div 86.4$ W, whereas the inlet concentration of ethanol in the stream of gases was in the range of $0.0028 \div 0.132$ mol/m³ ($0.128 \div 6.086$ g/m³).

The kinetic measurements were performed using the constant gas flow velocity \dot{F}_{0v} equal to 0.25 m³/h ($6.94 \cdot 10^{-4}$ m³/s). The calculated total residence time in the reactor τ_z' for these tests was of 1.39 s.

RESULTS AND DISCUSSION

The destructive oxidation of ethanol in the corona discharge reactor was carried out under isothermal conditions at the temperature of 303 ± 1 K. The results of gas chromatographic analysis of samples of the outlet mixture indicated that it did not contain intermediate products. It means that only CO₂ and H₂O were the final products of the reaction.

The measurements were performed in the wide ranges of the process parameters. First, they were analyzed using statistical methods. The Statistica 6.0 application was used for this analysis. The influence of inlet ethanol concentration in a stream of air, power supply to the corona reactor and the total residence time in the corona reactor on the

ethanol destruction are presented in graph in Figures 4 and 5.

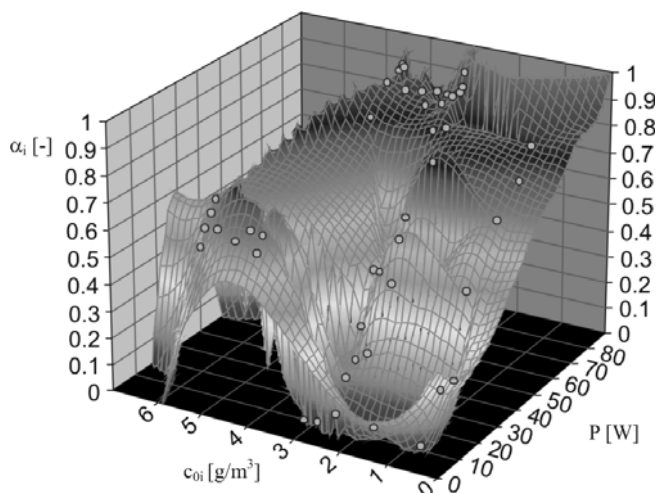


Figure 4. The influence of inlet concentration and power supply to the reactor on ethanol destruction

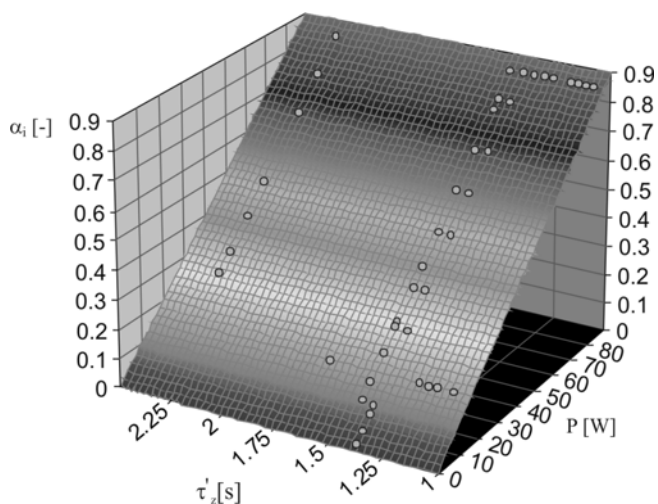


Figure 5. The influence of the total residence time of gases in the reactor and power supply to the reactor on ethanol destruction

The mathematical elaboration of these results allowed to derive the equations, which describe the ethanol destruction as a function of the reaction parameters.

In this way the destruction efficiency of ethyl alcohol can be described by the following functions of operation parameters:

– power supply to the reactor P and the inlet ethanol concentration in the stream of gases c_{0i} ,

$$\alpha_i = 0.3961 - 8.14 \cdot 10^{-3} P - 0.548 \cdot c_{0i} + 5.95 \cdot 10^{-4} P^2 + 0.207 \cdot c_{0i}^2 - 9.67 \cdot 10^{-4} P \cdot c_{0i} - 4.61 \cdot P^{-3} - 0.017 \cdot c_{0i}^3 - 6.69 \cdot 10^{-4} P \cdot c_{0i}^2 + 7.99 \cdot 10^{-6} P^2 \cdot c_{0i} \quad (R^2=0.936)$$

(11)

– power supply to the reactor P and the total residence time in the reactor τ_z' ,

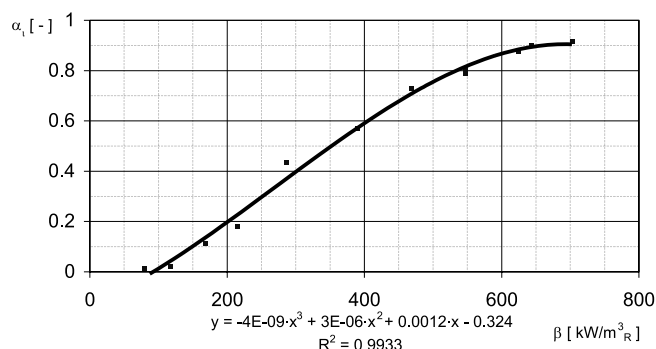
$$\alpha_i = -4.929 \cdot e^{-17} + 0.01 \cdot P + 4.958 \cdot e^{-17} \cdot \tau_z' + 2.629 \cdot e^{-21} \cdot P^2 - 1.124 - 2.434 \cdot e^{-19} \cdot P \cdot \tau_z'$$

(12)

Table 1. The results of the measurements and calculations for kinetic investigations

| No. | Input concentration c_{0i} [mol/m ³] | Output concentration c_i [mol/m ³] | Degree of destruction α_i [-] | Input high-voltage U [V] | Current I [A] | Power supply P [W] | Factor $\beta = P/V_R$ [kW/m ³ _R] |
|-----|--|--|--|----------------------------------|-----------------------|----------------------------|--|
| 1 | 0.06383 | 0.06306 | 0.012 | 140 | 0.070 | 9.8 | 79.675 |
| 2 | 0.05982 | 0.05850 | 0.022 | 160 | 0.090 | 14.4 | 117.073 |
| 3 | 0.06126 | 0.05421 | 0.115 | 180 | 0.115 | 20.7 | 168.292 |
| 4 | 0.05687 | 0.04657 | 0.181 | 240 | 0.110 | 26.4 | 214.634 |
| 5 | 0.05972 | 0.03380 | 0.434 | 320 | 0.110 | 35.2 | 286.179 |
| 6 | 0.05985 | 0.02577 | 0.569 | 400 | 0.120 | 48.0 | 390.244 |
| 7 | 0.05548 | 0.01503 | 0.729 | 480 | 0.120 | 57.6 | 468.293 |
| 8 | 0.06496 | 0.01371 | 0.789 | 560 | 0.120 | 67.2 | 546.341 |
| 9 | 0.05815 | 0.01122 | 0.807 | 560 | 0.120 | 67.2 | 546.341 |
| 10 | 0.06437 | 0.00804 | 0.875 | 640 | 0.120 | 76.8 | 624.390 |
| 11 | 0.06141 | 0.00620 | 0.899 | 720 | 0.110 | 79.2 | 643.902 |
| 12 | 0.06737 | 0.00573 | 0.915 | 720 | 0.120 | 86.4 | 702.439 |

The results of the measurements and the calculations for kinetic investigations (for constant $\dot{F}_{ov} = 0.25 \text{ m}^3/\text{h} = \text{constant}$) are presented in Table 1. On the basis of the experimental results the degree of ethanol destruction for different values of power supply to the reactor was calculated and the relationship between the degree of ethanol destruction and factor β ($\beta = P/V_R$, power consumption by unit of the active volume of the reactor) was determined (Figure 6). The dependence $\alpha_i = f(\beta) = f(P/V_R)$ was obtained by an approximation of measurement results by the third order equation.

**Figure 6.** Ethanol destruction degree as a function of factor β
 $\alpha_i = f(\beta) = f(P/V_R)$

It can be seen that a high ethanol destruction is obtained ($\alpha_i > 0.9$) when factor β has the value about 650 kW/m^3 .

On the basis of the results shown in Table 1, using equation (7), the values of function Z (apparent reaction rate constant) for different power supply to the reactor were also calculated. The dependence $\ln(Z) = f(1/P)$ is shown in Figure 7.

The approximation of results presented in Figure 7 gives a linear equation in the form:

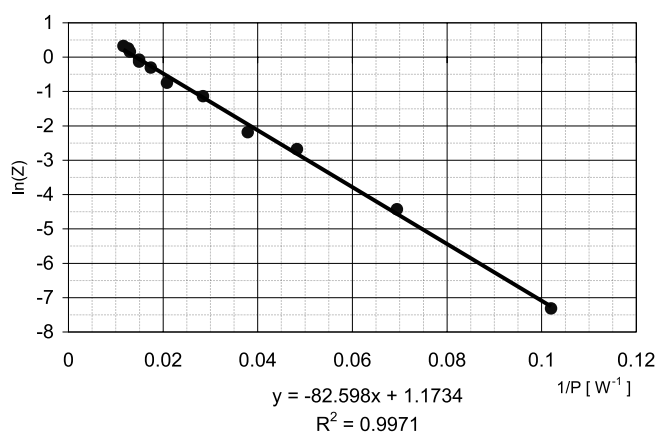
$$y = -82.598 \cdot x + 1.1734 \quad (R^2 = 0.9971)$$

From this equation the parameters A and B of equation (4) can be calculated

$$A = \exp(1.1734) = 3.233 \quad B = -82.598$$

Taking into account these values, the equation (4) can be written as:

$$Z = \varphi(P) = 3.233 \cdot \exp\left(-\frac{82.598}{P}\right) \quad (13)$$

**Figure 7.** The dependence of $\ln(Z) = f(1/P)$

After substitution of equation (13) into equation (5), the total rate of destructive ethanol oxidation in the corona discharge reactor used in the investigations can be described by the equation in the form:

$$r_i = 3.233 \cdot \exp\left(-\frac{82.598}{P}\right) \cdot c_{0i} \cdot (1 - \alpha_i) \quad (14)$$

This empirical equation describing the process rate can be used to design the corona reactor for the purification of the air or exhaust gases polluted with ethanol, when the concentration of this pollutant is in the range of $0.0028 - 0.132 \text{ mol/m}^3$ ($0.128 \div 6.086 \text{ g/m}^3$) and the space velocity of gases is in the range of $1220 \div 2680 \text{ m}^3/(\text{m}^3_R \cdot \text{h})$.

CONCLUSIONS

The following conclusions can be drawn from this work:

- High efficiency of the destructive oxidation of ethanol ($\alpha_i > 0.9$) is achieved in the corona discharge reactor at low temperature of 303 K and power consumption by unit active volume of the reactor about 650 kW/m^3_R . Only harmless CO_2 and H_2O were the final products of the reaction.

- The efficiency of ethanol destruction mainly depends on the power supply to the reactor and the input concentration of ethanol in stream of air.

- The reaction rate for the applied conditions of experiments i.e. inlet ethanol concentration in the stream of air in the range of $0.0028 - 0.132 \text{ mol/m}^3$ ($0.128 \div 6.086 \text{ g/m}^3$), the gas flow velocity in the range of $0.15 - 0.33 \text{ m}^3/\text{h}$

(space velocity of gases is in the range of $1220 \div 2680 \text{ m}^3/(\text{m}^3_{\text{R}}\cdot\text{h})$, can be described by the first order equation in relation to the ethanol concentration.

– Under isothermal conditions, the apparent reaction rate constant mainly depends on the power supply to the reactor (equation 13). The real reaction rate is well described by equation (14) and can be useful in design of the corona reactors for the purification of the air and industrial waste gases polluted with ethanol.

Nomenclature

| | |
|---------------------------|--|
| A, B | – parameters of equation, s^{-1} |
| c_{0i} | – inlet ethanol concentration, mol/m^3 or g/m^3 |
| c_i | – outlet ethanol concentration, mol/m^3 |
| D | – inner diameter of external pipe, m |
| d | – outer diameter of internal pipe, m |
| d_i | – inner diameter of internal pipe, m |
| $d\alpha_i$ | – differential of conversion degree |
| dV_R | – element of reactor volume |
| f | – frequency of applied voltage, Hz |
| \dot{F}_{0v} | – gas flow velocity, m^3/s , m^3/h |
| \dot{F}'_{0v} | – space velocity, $\text{m}^3/(\text{m}^3_{\text{R}}\cdot\text{s})$ |
| i, j, k | – index |
| I | – current, A |
| m | – size of set of active zones between external and internal pipes of the reactor |
| n | – size of set of inactive zones between external and internal pipes of the reactor |
| L | – length of reactor, m |
| L' | – length of reactor active zone, m |
| L'' | – length of reactor inactive zone, m |
| P | – power supply to the reactor, W |
| R^2 | – correlation coefficient, [-] |
| r_i | – rate of reaction, $\text{mol}/(\text{m}^3\cdot\text{s})$ |
| T | – temperature, K |
| U | – applied input high-voltage, V |
| V_R | – active volume of reactor, m^3 |
| Z | – apparent reaction rate constant, s^{-1} |
| α_i | – ethanol destruction degree, |
| β | – factor, $\text{kW}/\text{m}^3_{\text{R}}$ |
| $\phi(P), \psi(\alpha_i)$ | – auxiliary functions |
| τ_z | – total residence time in the reactor, s |
| τ_{zn} | – residence time in the reactor inactive zone, s |
| τ_{zr} | – residence time in the reactor active zone, s |

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