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Catalytic ignition, extinction and combustion of acetone in air on platinum wires of different diameters

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The research paper deals with the I/d ratio, based on the analysis of the analytical dependence of the stationary temperature of the catalyst wire on the heating current. It enables to estimate the range of current strength for a certain diameter of the catalyst wire, in which it is possible to observe the ignition and extinction of a heterogeneous catalytic reaction. The experimental studies for the catalytic oxidation of acetone in air on a platinum catalyst wire with diameters of 20, 70, and 95 μ m showed that, in case of considering convection this ratio as I/d·Nu^{1/2} should be corrected. As a result, there is a coordination of low- and high-temperature oxidation regimes, as well as critical conditions for catalytic ignition and quenching in the representation of the catalyst wire temperature from this complex. This assessment is possible in the case of utilizing an electrical scheme for heating the wire in the mode of current stabilisation.

Keywords: catalytic oxidation, platinum wire, diameter, acetone vapour.

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Introduction

Catalytic reactions on platinum offer an alternative and energy-efficient way to reaction products, increasing the speed or completeness of chemical transformations that are normally extremely slow [1]. Platinum is the most common catalyst for the complete oxidation of volatile organic compounds [2]. The catalyst surface is of a vitalimportance, so the addition of platinum nanoparticles to the surface of an oxide catalyst significantly accelerates the oxidation of a compound [2-5].

Combustible gas mixtures and flammable vapours are present in the working environment of many industries. This feature presents a key role for detecting their presence in a gas environment, as it is essential to reduce the potential hazard before this mixture reaches an explosive concentration [6]. The principle of a thermocatalytic sensor is based on the change in temperature of a sensitive element, such as a microtile, during the catalytic combustion of gas on its surface [7].

The catalysts in the form of thin metal wires are rarely used in scientific research. For instance, a copper catalyst in the form of a wire with a diameter of 180 mcm was used in the reaction between polylactic acid (PLA) and coumarin as presented in this research paper [8].

Platinum wire is the basis of the pelistor. The research paper [9] investigates the response of a catalytic sensor based on a platinum group metal wire with a diameter of 10 mcm. The heating of such a pelistor was accompanied by a pre-programmed change of the voltage on it, during which the hysteresis of the sensor's response to hydrogen vapour was observed. The response of the sensor sensitivity is explained by the kinetics of the hydrogen oxidation reaction. In case of low supply voltage, the kinetic mode of response is implemented, and the diffusion mode occurs at high supply voltage. The hysteresis peculiarities on different metals are explained by the partial conversion of metal oxides to the metal phase at temperatures above 500 °C and the reverse oxidation of the metal happens at temperatures below 400°C.

The experimental studies of the ignition of a fuel/oxygen/nitrogen mixture on a 127 mcm diameter platinum wire in a flow (with a low Reynolds number range) showed that the reaction of the catalytic ignition temperature with increasing fuel concentration differs in

quality. The ignition temperature of depleted propane/air and butane/air mixtures decreases with increasing fuel concentration in them, while the opposite trend is observed for depleted mixtures of propylene, ethylene, carbon monoxide and hydrogen with air [10].

The electrical scheme for heating is principal when using wires. The easiest way to implement the scheme is the non-isothermal catalytic oxidation of gases. The resistance of the wire, and subsequently its average temperature, depends on the operating parameter in the mode of stabilising the heating current or voltage. The most famous example is Buben's results on the nonisothermal oxidation of ammonia and hydrogen on a 100 mcm platinum wire in the mode of current stabilisation [11], which were repeated for acetone as well [12-13]. The catalytic ignition of gas impurities occurred when the current strength was increased to 0.6-0.75 A. In general, the dependence of the wire temperature on the current strength showed a hysteresis character. The influence of the wire diameter on the characteristic values of the catalytic ignition current of gases at that time did not rise

The similar studies of hysteresis during the oxidation of simple hydrocarbons (methane, ethane, propane, isobutane, ethylene, propylene) on a platinum plate were conducted by Veser and Schmidt [14], mixtures of ethane and carbon monoxide by Harold and Luss [15], and hydrogen by Chen and Song [16].

The heating mode of the platinum wire in [17] is isothermal, i.e., with the catalyst maintaining a stationary temperature. A quartz plate was used to stabilize and maintain temperature of 100 mcm.

During the oxidation of methane and ethane on platinum wires with diameters of 76, 203 and 250 mcm, the input heating power was controlled by the programmein the mode of stabilising the wire temperature. In a certain range of input power, each value corresponds to three different stationary values of the wire temperature [18]. This mode ensures a low probability of combustion transition to the gas phase.

Non-isothermal wire heating can be performed in the mode of voltage, power or current stabilization. The heating of the catalyst is proportional to the concentration of the combustible gas impurity. As a result, the catalytic combustion of gases can be used to convert harmful organic substances into safer ones. At first glance, the choice of scheme may seem insignificant. However, a more optimal scheme with current stabilisation leads to a simpler analytical description when using the effective temperature of the gas mixture [19]. In this case, the effective temperature gains weak dependence on the catalyst temperature.

The reaction of catalyst wires of different diameters in an environment with a vapour (gas)/air mixture is not being extensively studied.

The aim of this research paper is to determine the correlation between current strength and diameter, which will allow to estimate the range of current strength for the catalytic oxidation of organic compound vapours on a platinum wire of different diameters.

I. Analysis of the analytical dependence

The physical and mathematical model of heat and mass transfer of a long and thin metal wire is considered [20]. It is heated by an electric current in an electric circuit with stabilised current strength. The catalytic oxidation reaction of a small impurity of combustible gas in the air stream takes place on its surface. The condition of stationery state of the catalyst temperature in an air stream with a constant proportion of combustible gas impurities can be represented as a correlation of the current strength and the stationary temperature of the wire:

$$I = I_0 \sqrt{\frac{T - T_g}{T_0} - \frac{Y_f}{1 + Se} \frac{T_Q}{T_0} \frac{k_0}{\beta} \exp\left(-\frac{E}{RT}\right)},\tag{1}$$

$$\begin{split} I_0 &= \sqrt{\frac{\pi^2 d^2 \lambda_g N u T_0}{4\eta}}, \quad T_Q = \frac{Q_f L e}{c_{pg}}, \quad L e = \frac{D_f c_g \rho_g}{\lambda_g}, \\ k &= k_0 \exp\left(-\frac{E}{RT}\right), \quad S e = \frac{k}{\beta}, \quad \beta = \frac{D_f S h}{d}, \\ \eta &= \eta_0 (1 + \gamma (T - T_0) - \delta (T - T_0)^2), \end{split}$$

where Q_f – the thermal effect of the reaction, calculated per 1 kg of combustible gas, J/kg; Y_f – the relative mass fraction of the combustible gas impurity in the air; c_{pg} - is the isobaric specific heat of the gas mixture; T_g - is the temperature of the gas mixture; k – is the reaction constant in m/s; β – is the mass transfer coefficient of the combustible gas in m/s; η is the resistivity of platinum; D_f – is the diffusion coefficient of the combustible gas in air; c_g , ρ_g – are the specific heat capacity and density of the gas mixture; *Le*, *Nu*, *Sh* – are the Lewis, Nusselt, and Sherwood numbers.

The dependence I(T) has a minimum and a maximum at certain parameter points. The inverse dependence is hysteresis [20, 21]. At certain points of the current strength, there can occur catalytic ignition or extinction. For an analytical description of the conditions of catalytic ignition and quenching, the effective ambient temperature is introduced for effective operation [20]:

$$T_{g*} = T_g + \left(\frac{I}{I_0}\right)^2 T_0.$$
⁽²⁾

The second term of the right-hand side of (2) is the ratio of the heat generation power of the catalyst wire under current flow *P* and the product of the wire side surface area πdL and the heat transfer coefficient α . The product I/d and λ_g/η appears in the expression (2)when using an electrical circuit with a stabilized current (then $P = I^2 R = 4I^2 \eta L/d^2$). The latter relation is a weak function of the catalyst temperature and is independent of the wire length.

Then the critical condition for catalytic ignition can be presented as:

$$\frac{\Gamma_{Q}Y_{fi}E}{RT_{g*i}^{2}}\frac{k_{0}d}{D_{f}Sh}exp\left(-\frac{E}{RT_{g*i}}\right) = \frac{1}{e}i$$

$$T_{g} + \left(\frac{I_{i}}{I_{0}}\right)^{2}T_{0} = T_{i} - \frac{RT_{i,e}^{2}}{E}$$
(3)

The value of I_0 is a weak decreasing function of temperature. At the same time, it is proportional to the wire diameter $I_0 = d \cdot A$, where A is a function of temperature. Therefore, the stationary condition can be presented in the form:

$$\frac{I}{d} = A \cdot \sqrt{\frac{T - T_g}{T_0} - \frac{Y_f}{1 + Se} \frac{T_Q}{T_0} \frac{k_0}{\beta} \exp\left(-\frac{E}{RT}\right)}.$$
(4)

The main assumption that was made from the analysis of (2) and (4) is the possibility of the I/d invariant in the description of the catalytic oxidation of small gas impurities. For wires of different diameters with the same I/d, the value of the effective temperature of the gas medium T_{g*} is the critical temperature of catalytic ignition T_i is the same

In addition, at low-temperature catalytic oxidation (Se < 0.1) and high-temperature catalytic combustion (Se >> 1), dependence (4) is as follows

$$\frac{l}{d}\Big|_{Se<0.1} = A \cdot \sqrt{\frac{T-T_g}{T_0}} \quad \text{i} \quad \frac{l}{d}\Big|_{Se\gg1} = A \cdot \sqrt{\frac{T-T_g}{T_0} + \frac{T_Q}{T_0}Y_f}.$$

Thus, in these modes, I/d is a function of the catalyst temperature only.

Therefore, this assumption will further be experimentally verified.

II. Experimental plant.

The experimental studies were performed on the plant described in [19, 20]. The main component of the reaction chamber is a manometric thermocouple transducer PMT-2. After cutting off the glass flask from the lamp base, a platinum wire of the required diameter in the form of a snake was fixed to the electrodes in a plane perpendicular to the lamp axis. The hole was drilled in the lamp base through which the gas-air mixture was fed. The glass flask was then glued to the base. Platinum wires with diameters d of 20, 70, and 95 mcm were used for the study. Their length *L* was different, but the ratio is L/d > 500.

The gas mixture with combustible liquid vapours was created by passing air through the liquids (acetone or ethanol) in the flask. This flow could be diluted by passing clean air in parallel. The total flow rate was measured from the cold mixture.

The mixture was passed through a tube furnace. To ensure a constant temperature on the walls of the reaction chamber, the air stream was blown from the outside between the lamp bulb 1 (diameter 30 mm) and the outer chamber 10 (diameter 60 mm) (Fig. 1). This also enabled to reducesignificantly the difference in the temperature of the mixture before it was fed into the chamber and inside it (due to natural convection) to 20-30 °C in the catalytic combustion mode. The measurements (taking of the CVR) were carried out after the readings of both thermocouples 4 and 5 were established.

The platinum wire was connected to a DC power supply and a Picotest 3510 A multimeter in a three-wire circuit. The primary experimental data is the volt-ampere characteristic of the wire in the current stabilization mode. The contacts of the lamp have a diameter of 1 mm and their total resistance is $R_k = 0.105$ Ohm. It should be mentioned that the total resistance practically does not change in the range of currents for this research. The lamp contacts are 10 times thicker than the platinum wire. Therefore, heat dissipation through them is neglected.



Fig. 1. Schematic of the experimental plant: 1 - PMT-2 glass flask with interchangeable platinum wires (3) of different diameters, 2 - thick metal contacts; 4, 5 - millivoltmeters with HA thermocouples; 6, 7 - float-type flow meter, 8 - liquid container; 9 - air compressor; 10 - external chamber with air flow.

The quadratic dependence of the wire resistance on temperature was used to calculate the average wire temperature:

$$R_{c} = \frac{U}{I} - R_{k}, \quad \frac{R_{c}}{R_{c0}} = 1 + \gamma (T - T_{0}) - \delta (T - T_{0})^{2},$$
$$R_{c0} = \eta_{0} \frac{4L}{\pi d^{2}}.$$

In this formula $\gamma = 39.7 \cdot 10^{-4} \text{ 1/K}, \delta = 5.841 \cdot 10^{-7} \text{ 1/K}^2, \eta_0 = 9.8 \cdot 10^{-8} \text{Om} \cdot \text{m}.$

The parameters of the selected platinum wires are shown in Table 1.

		Table. 1
Dimensions of Pt wires for research		
d, mcm	L, mm	R_{c0} ,Ohm
20	34	10.6
70	50	1.28
95	56	0.775

The resistance of a 20 mcm wire varied from 15 to 45 Ohm during heating and cooling, and for 95 mcm from 0.85 to 4 Ohm.

III. The analysis of the results

Fig. 2 shows the dependence of the temperature of platinum wire with a diameter of 20 and 95 mcm on the current strength. The current strength gradually increased to the point of catalytic ignition and then reduced.



Fig. 2. Dependence of the stationary temperature of the platinum catalyst wire on the current in an acetone-air mixture. Calculation according to (1).
a) diameter 20 mcm, acetone concentration *Y_a*: 1) 2.3%(◊); 2) 2.8% (●), 3) 3.1%(○).

b) 95 mcm diameter, acetone Y_a concentration: 1) 2.1%(Δ); 2) 2.5%(\bullet), 3) 3.3%(\circ).

According to the classical concepts, low concentrations of combustible gas in the mixture (up to 5 %) can be estimated through the difference in the stationary temperatures of high-temperature catalyst heating T_m and low-temperature oxidation T_{g^*} during the course of one reaction in the diffusion area:

$$Y_f = \frac{T_m - T_{g*}}{T_Q} = \frac{c_{pg}(T_m - T_{g*})}{Q_f Le}.$$
 (1)

The parameters for calculating analytical dependences for the oxidation of acetone on platinum were taken from [20]: $Q_f = 31.5$ MJ/kg, E = 84 kJ/mol, $k_0 = 3 \cdot 10^6$ 1/s, $D_{f0} = 0.109 \cdot 10^{-4}$ m²/s, Le = 0.586, m = 0, n = 1.82, $\rho_{g0} = 1.293$ kg/m³, $c_{g0} = 1005$ J/(kg·K).

The value of Nusselt's number Nu was estimated from the comparison of calculated and experimental data in the field of stationary low-temperature regimes. It was found to be Nu = 0.37 (for d = 20 mcm), Nu = 0.37 (for d = 70 mcm) and Nu = 0.55 (for d = 95 mcm). Hatton's [22, p.210] dependence [23] for the Nusselt's number in the case of mixed convection during cylinder heat transfer is given in [22, p.210]. In works [23, 24], the smallest *Nu* value is 0.35.

A qualitatively similar pattern is observed for platinum wires of different diameters. At a certain concentration of acetone Y_a in the gas mixture, a hysteresis of the steady-state temperature with the current strength is noticed. With an increase in the concentration of Y_a , the critical value of the current strength I_i for catalytic ignition also decreases, and is weaker than I_e for catalytic quenching. The temperatures of a catalyst wire with a diameter of 95 mcm at catalytic quenching are 50-100 K lower than for a 20 mcmwire.

Consequently, the values of the I/d complex for different catalyst diameters should be considered at first. The representative value of the current during catalytic ignition was chosen, with the aim of comparison, which is weakly dependent on the concentration of the combustible gas. Table 2 shows the experimental values obtained for acetone and ethanol vapours.

The obtained data show that when the diameter is changed by a factor of 5, this ratio remains of the same order: for a smaller diameter, it is 10-20 % lower.

Fig. 3a demonstrates the comparison of the stationary dependences for platinum wires of 20 and 95 mcm in the form of dependence (4). The rate of change of the temperature of the 20 mcm diameter wire in the catalytic combustion mode is higher than for 100 mcm. The attention should be drawn to the proximity of acetone concentrations in the mixture at which thermal hysteresis of catalytic oxidation is performed.

The critical conditions (point i– catalytic ignition, point e – extinction) are determined by applying the extremum condition $\partial I/\partial T = 0$ to dependence (4). As a result, the system of equations together with (4) is obtained:

$$\left(\frac{I}{d} \right|_{i,e} = A \sqrt{\frac{T_{i,e} - T_g}{T_0} - \frac{R T_{i,e}^2}{E T_0} \left(1 + S e_{i,e} \right)},$$
(5a)

$$Y_f = \frac{RT_{i,e}^2 \left(1 + Se_{i,e}\right)^2}{ET_Q}.$$
 (5b)

At the points of catalytic spontaneous combustion, at a distance from the degeneracy point, $Se_i < 0.1$. Therefore, for Se<<1, the joint solution of equations (5) leads to [20]

$$T_g + \left(\frac{I_i}{Ad}\right)^2 T_0 = T_i - \frac{RT_{i,e}^2}{E},$$

$$\frac{T_Q Y_{fi}E}{RT_{g*i}^2} \frac{k_0 d}{D_f Sh} exp\left(-\frac{E}{RT_{g*i}}\right) = \frac{1}{e}.$$
 (6)

Under the condition of I/d values being the same, the critical value of the wire temperature during the catalytic ignition of T_i should be the same as well. However, as Fig. 3a allows to see, it does not correspond reality. The second condition (6) contains three important parameters: the effective temperature of the mixture, the diameter of the wire, and the mass fraction of fuel vapour. The value of Y_i has similar or nearer values, as it was mentioned above. Therefore, if T_{g^*} is assumed to have a stronger

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Table 2.

Experimental critical values of current obtained during catalytic ignition in air at a temperature of $T_g = 295$ K



Fig. 3. Dependence of the stationary temperature of platinum wire on the complex a) I/d; b) $I/(d \cdot Nu^{0.5})$. Blue lines are 20 mcm, black lines are 95 mcm.

influence in the exponent, then $T_{g*i} \sim 1/ln(d)$ is at catalytic ignition. That is, the critical value of the effective temperature of the mixture increases with a decrease in the wire diameter. At the same time, Fig. 3a depicts that the result is the opposite.

Nonetheless, the hidden dependence on the diameter is Nu(d). Note that $I_0 \sim d \cdot Nu^{1/2}$. This correction is being used, and Fig. 3b represents the results as a function of the stationary temperature of the catalyst as a function of the complex $I/(d \cdot Nu^{\frac{1}{2}})$. Such a representation agrees not only with the low-temperature and high-temperature regimes, their slope, relative position at different concentrations, but also with the increase in the critical temperature of catalytic ignition with a decrease in the wire diameter. On the ground of this, the obtained experimental differences in the critical I/d values can be explained by the difference in heat transfer of wires of different diameters.

The critical parameters of catalytic ignition and extinction at different catalyst diameters are presented in Fig. 4. It can be seen that when the wire diameter changes by almost 5 times, the concentration interval at which catalytic ignition and extinction are observed does not change essentially. At the same time, the difference between the values of $(I/d)_i$ and $(I/d)_e$ is as smaller, as lower the concentration of the combustible gas. It is evident that with a further decrease in the gas concentration, degeneration of the critical conditions should occur [25]: the critical values of I_i and I_e virtually coincide.



Fig. 4. Dependence of the critical value of the square of the complex $I/(d \cdot Nu^{0.5})$ on the acetone concentration. Calculation according to (5). Points for $d = 20 \text{ mcm} (\blacktriangle, \Delta)$, 70 mcm (\blacksquare, \Box) , 95 mcm (\bullet, \circ) .

Conclusions

As a result, having analyses the analytical dependence, the I/d ratio was proposed to be used as an estimation parameter to assess the range of currents at which non-isothermal catalytic ignition and extinction of gas vapours in air is attainable or possible.

Under the condition of convection, the corrected $I/(d \cdot Nu^{0.5})$ complex should be used for relative comparison of results for wires of different diameters. The use of this complex is possible when using an electrical scheme for heating the catalyst wire in the current stabilization mode.

In this case, the obtained critical dependences describing the conditions of catalytic ignition and quenching do not depend on the length of the wire (provided that L >> d).

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Каталітичне займання, погасання та горіння ацетону в повітрі на платинових нитках різного діаметру

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В роботі на основі аналізу аналітичної залежності стаціонарної температури нитки каталізатора від сили нагріваючого струму запропоновано співвідношення I/d. Воно дозволяє оцінити діапазон сили струму для певного діаметру нитки каталізатора, в якому можливе спостереження займання і погасання гетерогенно-каталітичної реакції. Експериментальні дослідження для каталітичного окислення ацетону в повітрі на платиновій нитці каталізатора діаметром 20, 70 та 95 мкм показали, що при врахуванні конвекції необхідно скорегувати це відношення як $I/d \cdot Nu^{1/2}$. В результаті в представленні температури нитки каталізатора від даного комплексу спостерігається узгодження низько- і високотемпературних режимів окислення, а також критичних умов каталітичного займанні і погасання. Дана оцінка можлива у випадку застосування електричної схеми нагрівання нитки в режимі стабілізації сили струму.

Ключові слова: каталітичне окислення, платинова нитка, діаметр, пари ацетону.