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# Ignition and Self-Supporting Burning of Gas-Air Mixtures with Hydrogen Admixtures on Platinum Wire

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The proposed work describes analytical identification of hydrogen admixture concentration and catalyst temperatures limit values beyond which catalytic flameless steady combustion of gas-air mixtures at ambient temperature at platinum wires is observed. Effect of gas-air slip velocity upon considered values is shown. Initial platinum wire preheating temperatures required for catalytic ignition are determined.

**Keywords:** metallic oxidation catalysts, wire, hydrogen, heat and mass transfer hysteresis, self-ignition, fade, surface (flameless) combustion.

Experimental researches of platinum wire temperature dependence on heating amperage showed that steady catalytic combustion of ignited cool gas-air mixtures with hydrogen and ammonium admixtures may take place even after electrical power supply is switched off [1]. Catalytic combustion in such conditions is advantageous since gaseous mixture with combustible gases admixture does not require continuous heating with only initial heating of platinum wire beyond certain critical temperature (point of ignition) [2]. In this case initial temperature of wire should exceed ignition point but less that fade point and combustible gases content in gas-air mixture should exceed the fade concentration value.

A question arises, "What does critical fade concentration value depend on?" Obviously, it depends on conditions accompanying heat and mass exchange between catalyst particles with air-gas mixture, which, in their turn, depend on particle size and flow velocity. In other words, dependence should be established between critical fade point for combustible gas admixture on wire diameter and air-gas mixture flow velocity with known temperature.

Therefore, aim of proposed work consists in analytical identification of minimum critical fade concentration of combustible gas and appropriate platinum wire temperature, beyond which self-supporting combustion of cool gas-air mixture with hydrogen admixture will take place on catalytic wire at specified temperature of steady air-gas flow $T_{g,\infty}$.

The work contains an analysis of ignition processes

(initial thermal effect) and stable stationery combustion of cool gas-air mixtures with combustible gas (hydrogen) admixture ($H_2 + 0.5O_2 \xrightarrow{p_t} H_2O$) on platinum wire. Heat exchange between pre-heated wire and cool air-gas mixture is described by Newton-Richmann principle represented by formula below,

$$q_h = \alpha(T - T_{g,\infty}) = \frac{\lambda_g Nu}{d}(T - T_{g,\infty}),$$

$$\delta_h = \frac{d}{Nu}, \quad \lambda_g = F(Y_i, T),$$

with $\lambda_g = F(Y_i, T)$ being a non-additive function of substances concentrations composing the mixture. Heat transfer coefficient experimental data validity is also related with heat losses due to radiation. More valid data may be obtained for mass transfer coefficient which is determined by diffusion concentrations factor. Combustion gas mass transfer to wire surface principle is represented, as follows

$$-j_{a,\beta} = \beta\rho_g(Y - Y_{a,\infty})$$

$$\beta = \frac{D_a}{\delta_h}, \delta_h = \frac{d}{Sh}, \quad Sh = 0.99 \, \mathrm{Re}^{0.305} Sc^m$$

$$(m = 0.33), \quad \mathrm{Re} = \frac{Ud}{\nu_g}, \quad Sc = \frac{\nu_g}{D_g},$$

with Re, Sc – Reynolds and Schmidt numbers, respectively, $U$ – flow linear velocity, $\nu_g$ – mixture kinematic viscosity, m²/s.
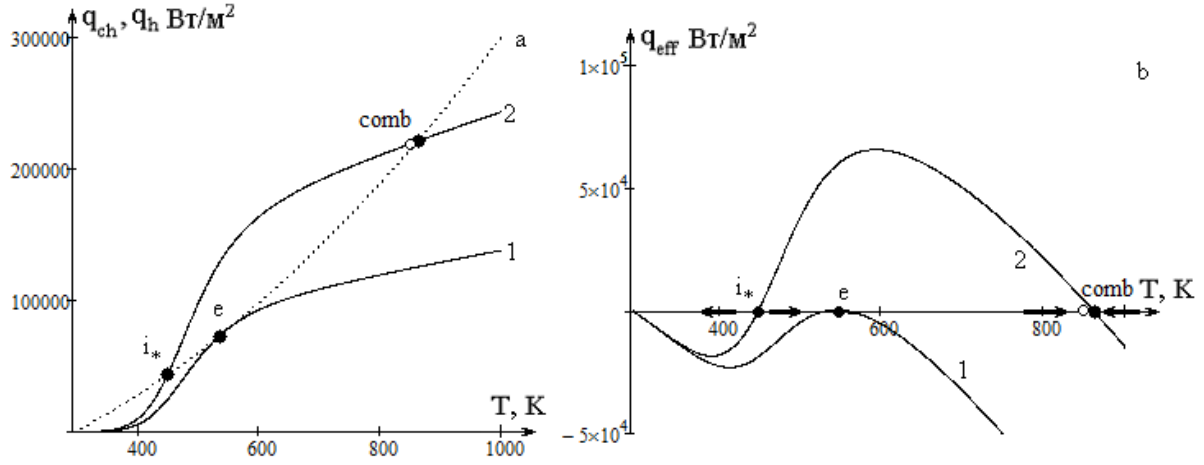
**Fig. 1.** Illustration of fade  1) $Y_{a,e} = 0.108\%$, and self-supporting combustion 2) $Y_a = 0.19\%$. a) Dependence between heat release rate $q_{ch}$ (solid line) and heat transfer $q_h$ (dotted line) and  b) efficient heat release rate  $q_{eff}$ for platinum wire with 100 mcm diameter placed in air-gas mixture at $T_g = 293$ K. ○ – data [1]. $T_{i*} = 448$ K, $q_{ch,i*} = q_{h,i*} = 43500$ Wt/m³; $T_e = 544$ K, $q_{ch,e} = q_{h,e} = 77000$ Wt/m³; $T_{comb} = 850$ K, $q_{ch,comb} = q_{h,comb} = 22000$ Wt/м³.

Therefore, Lewis analogy is feasible to be applied to describe heat exchange between the heated wire and cool air-gas mixture with admixtures

$$q_h = \alpha(T - T_{g,\infty}) = c_{p,g}\rho_g \beta Le^{-1+m}(T - T_{g,\infty}).$$

The heating of wire by means of electric current may be noted by means of gas-air mixture efficient temperature

$$q_h = \alpha(T - T_{g,\infty}) + \frac{I^2 R_{el}}{\pi d L} = \alpha(T - T_g),$$

$$\eta = \eta_*\left(1 + \xi(T - T_*)\right), \qquad (1)$$

де $\eta$ – platinum specific impedance, Om·m; $I$ – amperage, A

Transforming formula (1), the formula below is obtained

$$q_h = \alpha(T - T_g) = c_{p,g}\rho_g \beta Le^{-1+m}(T - T_g)$$

Efficient air-gas mixture temperature $T_g$ means a thermal value to which inert wire heated by electric current strives [3]:

$$T_g = T_{g,\infty} + A_I \left(\frac{2I}{\pi d}\right)^2, \quad A_I = \frac{\eta}{ShD_g\rho_g c_{p,g} Le^{-1+m}}$$

Representing the wire thermo dynamical equation taking into account hydrogen thermal diffusion transfer of oxidation catalyst wire as an opened system similar to particle motion dynamics equation, formula below is obtained:

$$c_c \rho_c \frac{V_c}{S_c}\frac{\partial T}{\partial \tau} = q_{eff}, \quad q_{eff} = q_{ch} - q_h, \quad T(0) = T_b, \quad (2)$$

$$q_{ch} = Q_a k \rho_{g,s} \frac{Y_a\left(1 + \psi_T\right)}{1 + Se},$$

$$k = k_0 \exp\left(-\frac{E}{RT}\right), \quad Se = \frac{k \cdot \rho_{gs}}{\beta \cdot \rho_g}, \quad B_{T_*} = \frac{\psi_T}{1 + \psi_T}\frac{T_*}{T},$$

$$\psi_T = (2 - n)\frac{T - T_{g,\infty}}{T} Le^m,$$

with $T_g$, $T_{g\infty}$ – efficient and actual air-gas mixture, respectively K; $Y_a$ – mass share of combustible gas admixture; $T$ – wire temperature, K; $q_{ch}$, $q_h$ - heat release and heat transfer rates, respectively Wt/m² ; $E$ – activation energy, J/mole; $R$ – universal gas constant, J/(mole K); $Se$ – diffusion and kinetic relation; $\psi_T$, $B_{T*}$ – correction for thermal diffusion; $Q_a$ – reaction heat effect, J/kg$a$; $V_C / S_c$ – relation between catalyst volume to surface area, m.

Analysis of $q_{eff}(T, Y_a)$ (Fig. 1a) enables to draw to a conclusion, that initial catalyst temperature $T(0) = T_b$ should exceed critical initial ignition point $T_{i*}$ (т. $i_*$ (Fig. 1) with admixture concentration $Y_a$, which should meet the requirement to breach the stability condition ( $q_{eff} = 0$, $\partial q_{eff}/\partial T > 0$ ) to provide steady self-supporting catalytic combustion of cool air-gas mixture.

Calculations of catalytic combustion of hydrogen were performed for following parameters' values: Platinum wire diameter 100 mcm, $E = 55$ кJ/mole, $k_0 = 0.8 \cdot 10^6$ m/s, $Q_a = 120.9$ MJ/kgH₂, $D_{a0} = 0.6 \cdot 10^{-4}$ m²/s, $m = 0.33$, $n = 1.82$, $Sh = 0.51$ ($U = 0.15$ m/s), $T_0 = 293$ K, $\lambda_{g0} = 0.024$ Wt/m·K), $\rho_{g0} = 1.293$ kg/m³, $c_{g0} = 1005$ J/(kg·K).

Self-supporting combustion of cool air-gas mixture occurs with admixture concentration $Y_a$ and is described by combustion temperature $T_{comb}$ (Fig. 1, point "comb"),

meeting a stability condition for stationery states ($q_{eff} = 0$, $\partial q_{eff}/\partial T > 0$). Admixture concentrations $Y_a$ and steady combustion temperature $T_{comb}$ always exceed respective values of fade concentrations and temperature $Y_{a,e}$ and $T_e$, meeting the fade condition

($q_{eff} = 0$, $\partial q_{eff}/\partial T = 0$, $\partial^2 q_{eff}/\partial T^2 < 0$) (Point "e".Fig. 1a). Obviously, $Y_{a,e}$ depends on $d/Sh$ correlation.

Stationery dependences $T_g(T)$ та $Y_a(T)$ may be found from equation (3) below:

$$Q_a \rho_{gs} Y_a \frac{1+\psi_T}{1+Se} k_0 \exp\left(-\frac{E}{RT}\right) = c_{p,g} \rho_g \frac{Sh \cdot D}{d} Le^{-1+m}(T-T_g) \quad , \tag{3}$$

And may be represented, as follows,

$$T_g = T - \frac{Q_a \rho_{g,s} Y_a d k_0}{c_{p,g} \rho_g D_a Sh Le^{-1+m}} \frac{1+\psi_T}{1+Se} k_0 \exp\left(-\frac{E}{RT}\right) , \tag{4a}$$

$$Y_a = \frac{\rho_g}{\rho_{g,s}} \frac{1+Se}{1+\psi_T} \frac{Sh \cdot D}{d \cdot k_0} \frac{c_{p,g}(T-T_g)}{Q_a Le^{1-m}} \exp\left(\frac{E}{RT}\right) . \tag{4b}$$

Relations between self-ignition and fade concentrations $Y_a\big|_{i,e}$ in dependence on air-gas mixture temperature $T_g\big|_{i,e}$ may be applied to find fade concentration in parametrical format (see next page):

$$\begin{cases} Y_a\big|_{i,e} = \dfrac{c_{p,g} RT^2}{Q_a Le^{1-m} E} \dfrac{Sh \cdot D \cdot \rho_g}{d \cdot k_0 \cdot \rho_{gs}} \exp\left(\dfrac{E}{RT}\right) \cdot (1+Se)^2 \dfrac{(1-B_{T*})}{(1+\psi_T)}, & \tag{5a} \\[3mm] T_g\big|_{i,e} = T - \dfrac{RT^2}{E}(1+Se)(1-B_{T*}). & \tag{5b} \end{cases}$$

calculated in work [4] for a case of hydrogen thermocatalytic oxidation on platinum wire.

Equations system (5a) - (5b) may be obtained from (4) and i умови екстремумів на залежностях $T(T_g)$ (with $d$, $Y_a$, $Sh$ = const.) and $Y_a(T)$ (with $d$, $T_g$, $Sh$ = const).

Each of relations (4) determines a hysteresis area

(Fig. 2) in appropriate stationery relations $T_g(T)$ and $Y_a(T)$. Low temperature stationery conditions are located to the left from hysteresis area, high temperature stationery states (catalytic combustion of gases) are located to its right. Catalytic reaction should be triggered by means of ignition to achieve catalytic combustion modes subject to specific conditions (gaseous mixture
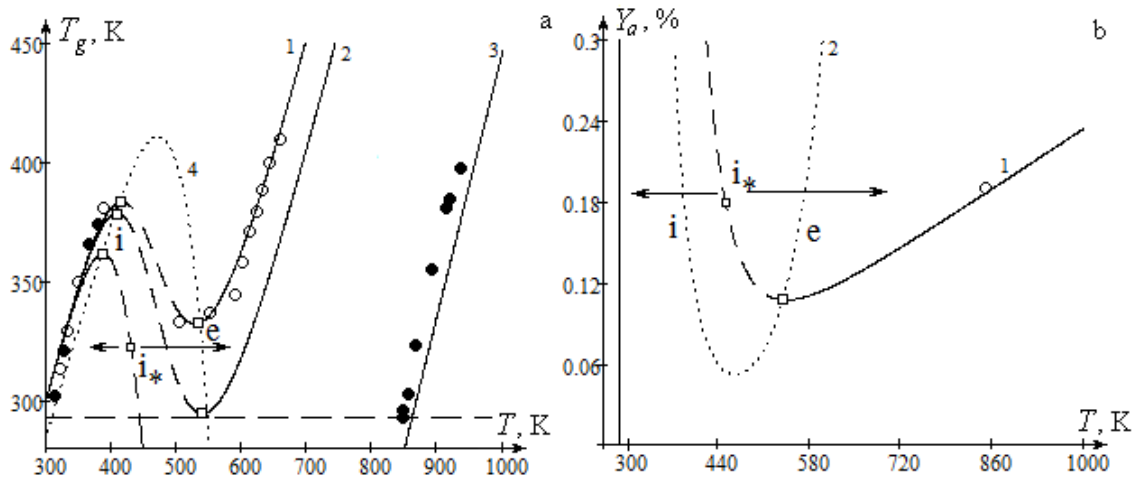


**Fig. 2a.** Relation between air-gas mixture with hydrogen admixtures and platinum wire stationery temperature at 1) $Y_a = 0.09\%$, 2) $Y_a = 0.108\%$, 3) $Y_a = 0.19\%$ ($d$ = 0.1 мм, $Sh$ = 0.51, $U$ = 0.15 м/с). Dotted parabolic line 4 – spinodal (5b).
**Fig. 2b.** Relation between hydrogen content in cool air-gas mixture ($T_g$ = 293 K) and platinum wire stationery temperature. ○ – experimental data. Dotted parabolic line 2 – spinodal (5a).

temperature, combustible gases ratio) meeting the hysteresis area parameters. It means that the initial catalysis temperature (platinum wire) should be increased to exceed ignition temperature, a certain critical value located between points **i** and **e**. Critical values of catalytic wire initial temperature (ignition point, Fig. 2 ) are located within $T_i < T_{i*} < T_e$, meeting the equation (3). Critical temperature and concentrations values are located within respective intervals, as below:

$$T_{ge} < T_{g,i*} < T_{gi}, \qquad Y_{a,i} < Y_{a,i*} < Y_{a,e}.$$

It should be noted that combustible gas ratio increasing, even in several times does not practically affect initial temperature critical value (Fig. 2a). It is explainable, since the ignition process runs mainly in kinetic mode of catalytic reaction and, thus, critical ignition value is a weaker function of combustible gas admixture ratio.

Analyzing (5a) – (5b) demonstrates that there is only one possible solution for $Se > 1$ meeting critical fade
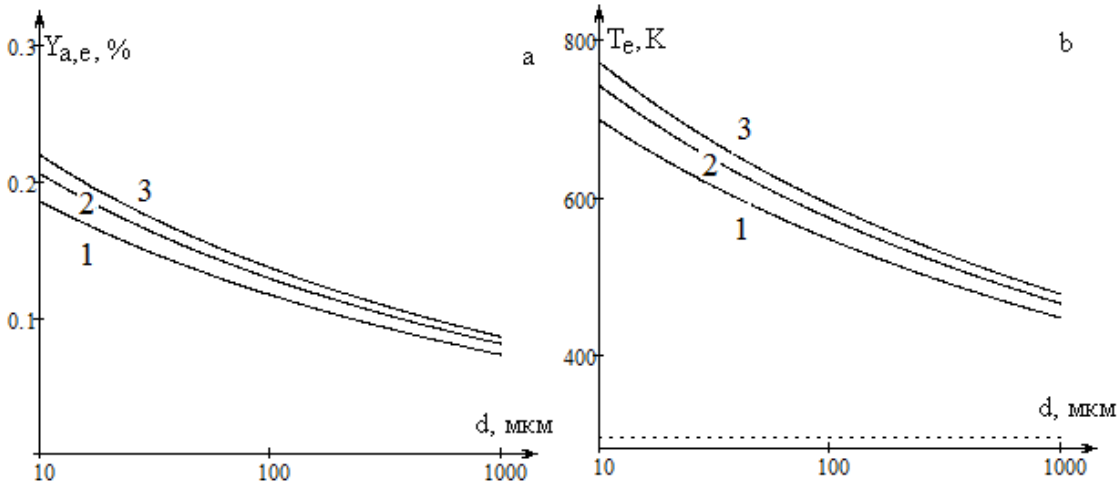
temperature value and combustible gas fade mass ratio $Y_{ae}$ at specified air-gas mixture temperature, e.g., $T_g = 293K$. Applying 5(b) diffusion and kinetic ratio and platinum wire diameter should be expressed, as follows:

$$Se_e = \frac{(T-T_g)E}{RT^2(1-B_{T*})} - 1, \qquad (6)$$

$$d_e = \frac{D_g Sh \rho_g}{k \rho_{gs}} \left( \frac{(T-T_g)E}{RT^2(1-B_{T*})} - 1 \right). \qquad (7)$$

Placing (7) into (5a), formula below is obtained

$$Y_{a,e} = \frac{c_{p,g}RT^2}{Q_a Le^{1-m}E} \frac{\left( \frac{(T-T_g)E}{RT^2} \right)^2}{\frac{(T-T_g)E}{RT^2} - (1-B_{T*})} \frac{1}{(1+\psi_T)}, \qquad (8)$$



**Fig. 3**. Relation of critical fade concentration $Y_{ae}$ (a) and critical fade temperature (b), beyond which stable flameless combustion takes place with electrical heating switched, depending on platinum wire diameter blown by an air-gas mixture at ambient temperature (293 K): 1) $U = 0.15$ m/s, 2) $U = 0.75$ m/s, 3) $U = 2$ m/s . Calculation applying (7), (8).
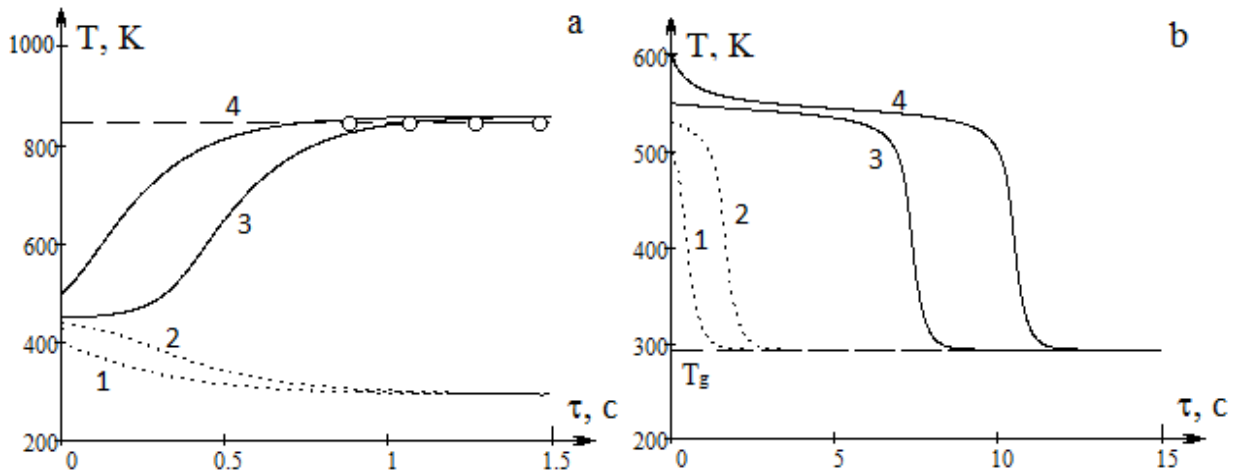


**Fig. 4.** Relation between platinum wire temperature in gaseous mixture at ambient temperature $T_g = 293K$ with hydrogen ratio a) $Y_a = 0.19$ %, b) $Y_a = 0.107$ % at time. Initial wire temperature: a) $T_{b1} = 400$ K, $T_{b2} = 440$ K, $T_{b3} = 450$ K, $T_{b4} = 500$ K. b) $T_{b1} = 500$ K, $T_{b2} = 530$ K, $T_{b3} = 550$ K, $T_{b4} = 600$ K.

Reviewing graph $Y_{a,e}(d)$ applying Sherwood criterion $Sh = 0.51$ (Fig. 3a), it becomes notable that adequate increase of platinum wire diameter enables to reduce $Y_{a,e}$ and, consequently, limit value of catalytic combustion $T_e$ (Fig. 3b). Fade temperature $T_e$ is a catalytic combustion limit temperature. Thermal diffusion reduces $Y_{a,e}$ additionally by 10-15%.

Thus, for a platinum wire with diameter $d = 0.1$ mm being run about with що mixture flow with ambient temperature at a linear speed of 0.15 m/s $Y_{ae} = 0.104\%$, which corresponds to hydrogen ratio 1.51 % by volume. This value meets the experimental range of hydrogen ratio 0.95-1.95% by volume, enabling self-supporting catalytic combustion with electrical supply off [2].

As it is obvious from (7) and (8), only critical catalyst diameter depends on Sherwood number and, appropriately, on flow velocity according to $d_e$ $U^{0.305}$ correlation. It is due to convective heat losses that critical hydrogen admixture ratio and catalyst temperature required for self-supporting catalytic combustion increase slowly, as the mixture flow velocity about the catalyst increases (Fig. 3).

Fig. 4a and 4b represent catalyst wire temperature in time with various hydrogen admixture concentrations in a mixture at ambient temperature with critical ignition i∗ point and fade point e (Fig. 1). For instance, with hydrogen ratio $Y_a = 0.19\%$ as an initial temperature increases over 450 K the air-gas mixture turns to catalytic combustion with catalytic combustion temperature does not depend on initial temperature exceeding the ignition point. With less hydrogen ratio $Y_a = 0.107\%$ (slightly less than critical fade concentration of mixture at ambient temperature) temporal dependence of wire temperature are also apparently different. With initial wire temperatures $T_b > T_e$ (graphs 3 and 4) catalytic

combustion is maintained for a substantial period with further running at low temperature mode. With $T_b < T_e$ (graphs 1 and 2) – the wire, as the time runs, cools down to ambient temperature. Different time of wire cooling down is explainable due to slow temperature change near critical fade point (minor values of efficient heat release rate Fig. 1).

## Conclusions

Thus, the represented system of parametrical equations (7) and (8), describing relation between $Y_{a,e}$, beyond which self-supporting catalytic combustion of air-gas mixture occurs, and platinum wire diameter at specified stationery temperature of cool air-gas flow with DC supply off (Fig. 3). It should be noted that ignition amperage or ignition point of air-gas mixture don't practically change as the combustion gas ratio increases. Lewis analogy between heat and mass transfer enables to calculate critical ignition and self-supporting combustion concentrations for air-gas mixtures with combustible gases admixtures with higher precision.

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# Запалювання та самопідтримуюче горіння газоповітряних сумішей з домішками водню на платиновому дротику

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В роботі аналітично знаходяться граничні значення концентрації домішки водню та температурі каталізатора, вище яких спостерігається каталітичне безполум'яне стаціонарне стійке горіння газоповітряних сумішей кімнатної температури на платинових дротиках. Показаний вплив швидкості обтікання га повітряної суміші на шукані величини. Визначені області початкових температур платинової нитки, до яких потрібно попередньо нагріти для здійснення каталітичного запалювання.

**Ключові слова:** металеві каталізатори окислення, нитка, водень, гістерезис тепломасообміну, самозаймання, погасання, поверхневе (безполум'яне) горіння.