V.V. Kalinchak[1], A.S. Chernenko[1], M.V. Sikorskyi[1], A.N Sofronkov[2], A.V. Fedorenko[1]

Cool Air-Gas Mixtures with Combustible Gas Admixtures Steady Flameless Combustion Delay Time on Platinum Particle (Wire)

[1]Chair of Thermal Physics, Odessa I. I. Mechnikov National University, Odessa, Dvoryanskaya str, 2, 65082, e-mail: chernalex@ukr.net
[2]Odessa State Ecology University, Odessa, Lvovskaya str, 15, 65000, teplophys@onu.edu.ua

The proposed work shows analytical way to find a build-up time (induction period) for a mode of steady flameless catalytic combustion on metallic wire for cool air-gas mixtures with combustible gaseous admixtures using as an example flameless combustion of air-gas mixtures with hydrogen admixture of a platinum wire. Steady flameless combustion is observable after an induction period due to increase of initial catalyst temperature exceeding critical point of ignition which depends on particle diameter and combustible gaseous admixture content. Total time of build-up time is split into two stages, being the time of catalytic reaction proceeding in transient and diffusion stages, respectively. Effects of combustible gas ratio, catalyst wire diameter and excess of initial temperature compared with point of ignition upon particular stages duration and induction period are illustrated.

Keywords: metallic oxidation catalysts**,** platinum wire, hydrogen, induction period, ignition, self-ignition, fading, surface (flameless) combustion.

Catalysts in disperse form play an exclusive role not only in nature, but also in the industry, where platinum metal platinum was widely used [1]. Catalytic combustion of gases proceeds directly on the solid catalysts's surface without open flames in the gas mixture. The work of the thermocatalytic gas analyzer [2] is based on the measurement of the amount of heat released in the non-flame burning mode. At the same time, it is necessary to know the time (induction period), through which the gas analyzer will enter the operating mode.

The work [3] experimentally demonstrates, that catalytic combustion of air-gas mixture with hydrogen or ammonium content on a surface of thin platinum wire becomes self-sustaining in steady stationery mode after electric current stoppage in a wire or after air-gas mixture temperature is reduced to any low temperature, provided the combustible gas admixture ratio exceeds a certain critical value of fading concentration at a specified temperature of cool air-gas mixture [4]. Under such conditions high-temperature catalytic oxidation of air-gas mixtures containing combustible gas is possible with initial temperature of catalyst wire exceeding a certain

critical value (catalytic ignition). Initial catalyst temperature critical value lies in an interval between catalytic self-ignition point and fade point of a specific air-gaseous mixture for a specified catalyst: $T_i < T_{b.i} < T_e$ [4].

The proposed work has its aim to identify time of delay for catalytic ignition of air-gas mixtures containing combustible gas with reaction on metallic wire (particle) proceeding as the first-order reaction.

I. Problem Statement

Catalyst metallic wire (particle) placed in stationery air-gas mixture containing combustible gas thermodynamic equation solutions should be reviewed. Here the assumptions, as below should be used,

1. Thermal resistance of the catalyst metallic wire (particle) is substantially less than that of air-gas mixture (the Biot number is substantially less than 1). It enables to ignore temperature distribution along the catalyst.

2. Thermal and concentration fields in gaseous phase are quasi-stationery, since the heat transfer factor for air-

gas mixture substantially exceeds that of metallic catalyst.

3. Thermal losses for radiation may be ignored due to minor size of the catalyst.

4. Heat and mass transfer between the catalyst particle and air-gas mixture is described by means of the Lewis analogy and Newton/Richmann principle (heat transfer by molecular and convective mechanisms q_h).

5. Chemical heat emission velocity rate q_{ch} and heterogenous reaction velocity at the catalyst particle surface depends on the first-order kinetics in view of admixture ratio and increase with the increase of temperature subject to the Arrhenius Principle.

In view of the above assumption the catalyst particle differential equation as an open system for a stationery air-gas flow will take a form, as follows,

$$c_c \rho_c \frac{V_c}{S_c} \frac{\partial T}{\partial \tau} = q_{eff}, \qquad q_{eff} = q_{ch} - q_h,$$

$$T(\tau = 0) = T_b, \qquad (1)$$

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

$$k = k_0 \exp\left(-\frac{E}{RT}\right), \quad \psi_T = (2-n)\frac{T-T_g}{T} Le^m,$$

$$Se = \frac{k \cdot \rho_{gs}}{\beta \cdot \rho_g}, \ \beta = \frac{D \cdot Sh}{d}, \ D = D_0 \left(\frac{T_*}{T_0}\right)^n, T_* = \frac{T + T_{g*}}{2},$$

with T_g – air-gas mixture temperature, K; Y_a – mass ratio of combustible gas admixture; T – wire temperature, K; q_{ch}, q_h – heat emission and heat extraction velocities, respectively, Wt/m^2; E – activation energy, J/mole; R – universal gaseous constant, J/(mole·K); Se – diffusion and kinetic ratio; ψ_T – thermal diffusion correction; Q_a – reaction heat effect, J/kg$a$; V_c / S_c – catalyst volume to surface area ratio, m; T_{g*} = 380 K – typical gaseous mixture temperature, c_c – catalyst specific heat capacity, J/kg K; ρ_c – catalyst density, kg/m^3. n – degree in the temperature dependence of the combustible gas's
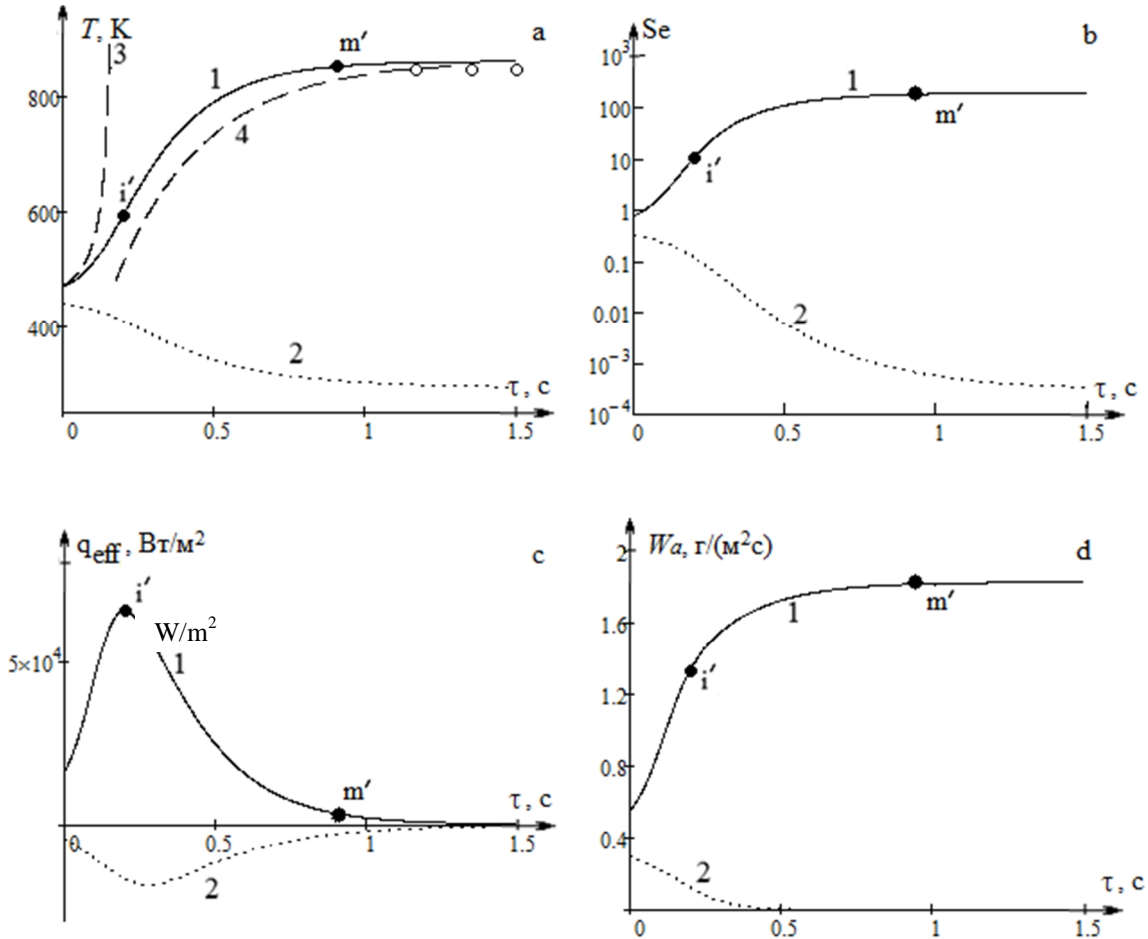

Fig. 1. Time characteristics of: a) platinum wire temperature, b) diffusion and kinetic correlation, c) efficient heat emission power, d) reaction speed for active combustible component in gaseous mixture with temperature T_g = 293K with hydrogen content Y_a = 0.19 %. Initial wire temperature: T_{b1} = 470 K, T_{b2} = 440 K. Initial temperature critical values: $T_{b.i1}$ = 448 K. Wire diameter 0.1 mm, Sherwood number Sh = 0.5. Specified diameter 0.2 mm. Ignition delay time (time as m' point is reached): τ_{ign} = 0.906 sec, o – experimental [3, P. 416]. Curves 1, 2 – solution (1), curve 3 – solution (9), curve 4 – solution (10).

diffusion coefficient, m – degree in dependence of the number of Sherwood on Schmidt number $Sh \sim Sc^m$ [3, C. 406].

Fig. 1a illustrates time function for catalyst wire temperature with different initial temperature values in air-gas mixtures of ambient temperature. Graphs 1 correspond to catalyst ignition and Graphs 2 to its absence. Boundary initial temperature T_{i*}, beyond which ignition occurs is identified as catalytic ignition temperature. Its values are found solving a non-stationery problem and are specified in notes to Fig. 1. Its value increases steadily as the combustible gas admixture ratio reduces. Time of ignition depends on difference between initial temperature and ignition temperature, the less is the difference the more is ignition time. Similarly, combustible gas admixture ratio decrease leads to increase in ignition delay time. Entire build-up period τ_{ign} to m' point (point m' correspond to the end of non-stationery combustion stage) to high-temperature catalytic oxidation mode may be split into two stages with the i' point (flex point on time curve of the catalyst temperature, $T_{m'} = 0.99 T_m$). In this case, the diffusion-kinetic ratio (the Semenov's number Se) is greater than one (Fig. 1b), and the chemical reaction's rate doesn't reach the steady-state value.

Parameters with following values have been applied for calculations of a case of hydrogen catalytic combustion [5]: platinum wire diameter 100 mcm, $E = 55$ kJ/mole, $k_0 = 0.8 \cdot 10^6$ m/sec, $Q_a = 120.9$ MJ/kgH$_2$, $D_0 = 0.6 \cdot 10^{-4}$ m^2/sec, $m = 0.33$, $n = 1.82$, $Sh = 0.51$ ($U = 0.15$ m/sec), $T_0 = 293$ K, $\lambda_{g0} = 0.024$ Wt/(m·K), $\rho_{g0} = 1.293$ kg/,m^3, $c_{g0} = 1005$ J/(kg·K), $c_c = 131$ J/(kg·K), $\rho_c = 21460$ kg/m^3.

II. Ignition point calculation

Review $q_{eff}(T, Y_a)$ is made for three specified platinum wire diameters to illustrate ignition conditions for air-gas mixture with hydrogen content (Fig. 2). Power

of chemical heat extraction in diffusion area has been used as a reference point.

$$q_{dif} = Q_a \beta \rho_g Y_a = Q_a \frac{Sh \cdot D}{d} \rho_g Y_a .$$

Obviously, that initial catalyst temperature $T(0) = T_b$ should exceed critical initial ignition point T_{i*} (point i_* Graph 2 Fig. 2) with admixture ratio Y_a meeting the breach of the stationery stability conditions.

Point i_*: $\qquad q_{eff} = 0, \quad \partial q_{eff} / \partial T \big|_{b,i} > 0$.

Combustion temperature T_m (Fig. 2, point m) meets the stationery conditions stability requirements:

Point m: $\qquad q_{eff} = 0, \partial q_{eff} / \partial T \big|_m < 0$.

Initial catalyst temperature at the stage of ignition (ignition point) should exceed the self-ignition temperature and less than fade point, see below $T_i < T_{b,i} < T_e$.

These limiting values meet the conditions of self-ignition and fading (points i and e):

$$q_{eff} = 0, \quad \partial q_{eff} / \partial T \big|_{i,e} = 0, \quad \partial^2 q_{eff} / \partial T^2 \big|_e < 0 ,$$

$$\partial^2 q_{eff} / \partial T^2 \big|_i > 0 .$$

and may be evaluated as

$$T_i \approx T_g + R T_g^2 / E \ \text{i} \ T_e = T_m - R T_m^2 / E ,$$

де $\quad T_m = T_Q + T_g , \qquad T_Q = \frac{Q_a Y_a}{c_g}(1 + \psi_T) Le^{1-m} ,$

$$\psi_T \approx (2-n)\left(1 - \frac{T_g}{T_g + Q_a Y_a Le^{1-m} / c_g}\right) Le^m .$$

Ignition point as a stationery temperature is obtained from the equation, as below

$$Q_a k_0 \exp\left(-\frac{E}{RT_{b,i}}\right)(1 + \psi_T)\rho_g Y_a = \beta c_g \rho_g Le^{-1+m}\left(T_{b,i} - T_g\right)\left[1 + \frac{k_0}{\beta}\exp\left(-\frac{E}{RT_{b,i}}\right)\right] .$$

Applying this equation mass exchange factor and specified diameters may be identified as a function of stationery temperature $T_{b,i}$

$$\beta = \left(\frac{T_m - T_{b,i}}{T_{b,i} - T_g}\right)k_0 \exp\left(-\frac{E}{RT_{b,i}}\right) \quad \text{and}$$

$$\frac{d}{Sh} = \left(\frac{T_{b,i} - T_g}{T_m - T_{b,i}}\right)\frac{D}{k_0}\exp\left(\frac{E}{RT_{b,i}}\right) . \tag{2}$$

d/Sh value is assumed as a specific diameter. Catalyst in the form of a sphere (particle) and a cylinder (wire) with

identical specific diameter possess equal ignition point characteristics as, well as other parameters.

Fig. 3 a, b represents obtained functions (2) in semi-logarithmic coordinates. They enable to obtain catalytic ignition temperature values for pre-given conditions. Point of ignition may change in broad range depending on mass exchange factor (catalyst diameter). In this case appropriate Semenov number values aren't small.

Semenov number is less than 1 $Se_{b,i} < 1$ for a substantial range of ignition points adjacent to T_i point. For ignition points range adjacent to T_e point Semenov number value exceeds 1: $Se_{b,i} > 1$.

Ignition point represented with formula below corresponds to $Se_{b,i} = 1$

$$T_{Q/2} = T_g + \frac{T_Q}{2}.$$

Limiting temperature's value is $T_{Q/2} = 575$ K for these parameters and the mass concentration of hydrogen $Y_a = 0.0019$. It corresponds to the given wire's diameter $d/Sh = 20.5$ мкм.

With $T_i < T_{b,i} < T_{Q/2}$ and $Se_{b,i} < 1$ relation (2) is practically exponential $\ln \beta \quad a_1 T + b_1$ (Fig. 3b), whereas with $T_{Q/2} < T_{b,i} < T_e$ and $Se_{b,i} > 1$ relation (2) is close to

exchange factor $\beta = 0.785$) ignition point critical value subject to (2) amounts to 448 K whereas, referring to (3a) correlation, it is 476 K.

Approximated relation, as below, may be applied to estimate ignition point within an area adjacent to fade point:

$$T_{b,i} \approx T_i + \left(T_{Q/2} - T_i\right) \frac{\beta - \beta_e}{\beta_{Q/2} - \beta_e}. \qquad (3, b)$$

Possibility to apply approximate formulae (3a) and (3b) may be observed using Figs. 2b and 2d, with Q/2 being a flex point.
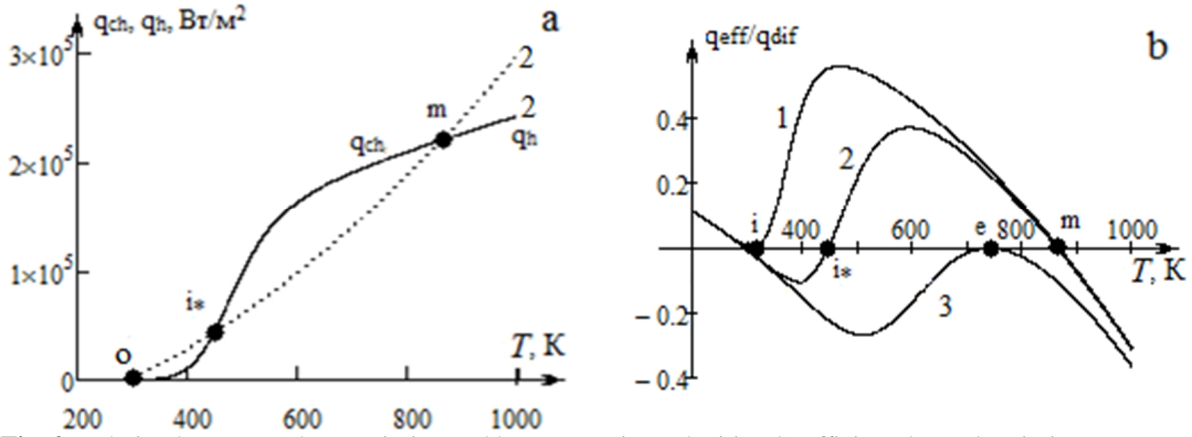

Fig. 2. Relation between a) heat emission and heat extraction velocities, b) efficient thermal emission power on platinum catalyst temperature in gaseous mixture at a temperature $T_g = 293$K with hydrogen ratio $Y_a = 0.0019$. Platinum catalyst specified diameters are: 1) 7.3 mm, 2) 0.2 mm, 3) 11 mkm. a) Solid curve is the chemical heat's rate $q_{ch}(T)$, dashed curve - heat transfer velocity $q_h(T)$, specific diameter 0.2 мм; b) Efficient thermal emission power $q_{eff}(T)/q_{dif}$, platinum catalyst's specific diameter: 1) 7.3 мм, 2) 0.2 мм, 3) 11 мкм.

linear $\beta \quad a_2 T + b_2$ (Fig. 3d).

So, approximated relation may be applied to estimate analytically ignition point in an area adjacent to self-ignition temperature.

$$T_{b,i} \approx T_i + \left(T_{Q/2} - T_i\right) \frac{\ln\left(\beta/\beta_i\right)}{\ln\left(\beta_{Q/2}/\beta_i\right)}. \qquad (3, a)$$

So, for platinum wire of 100 mkm diameter (mass

III. The first stage of ignition period

The problem (1) seems convenient to be reviewed in non-dimensional form. To begin with, heat exchange factor, expressed via stationery temperature, is placed into (1):

$$c_c \rho_c \frac{V_c}{S_c} \frac{\partial T}{\partial \tau} = Q_a k \rho_{g,s} \frac{Y_a\left(1+\psi_T\right)}{1+Se} - Q_a k_{b,i} \rho_{g,s} \frac{Y_a\left(1+\psi_T\right)}{1+Se_{b,i}} \cdot \frac{T-T_g}{T_{b,i}-T_g}, \quad T(0) = T_b, \qquad (4)$$

Values, applied by Frank-Kamenetskiy [3] are convenient to be chosen as non-dimensioned indicators of temperature and time. Here application of ignition point instead of the gaseous mixture temperature as characteristic temperature construes a specific feature:

$$\theta = \frac{T-T_{b,i}}{RT_{b,i}^2} E, \quad \theta_g = \frac{T_g - T_{b,i}}{RT_{b,i}^2} E.$$

$$\text{\%} = \frac{S_c \tau E}{c_c \rho_c V_c RT_{b,i}^2} Q k_0 \exp\left(-\frac{E}{RT_{b,i}}\right) \rho_g Y_a \left(1+\psi_T\right) \frac{1}{1+\dfrac{k_0}{\beta}\exp\left(-\dfrac{E}{RT_{b,i}}\right)}.$$

The resulting transformation represents a problem (4) in non-dimensional form:

$$\frac{d\theta}{d\varkappa} = \left(\frac{1+Se_{b,i}}{1+Se_{b,i}\exp\theta}\right)\exp\theta - \left(\frac{\theta}{|\theta_g|}+1\right),$$

$$\theta(\varkappa=0)=\theta_b, \qquad (5)$$

$$\bar{q}_{ch} = \left(\frac{1+Se_{b,i}}{1+Se_{b,i}\exp\theta}\right)\exp\theta, \quad \bar{q}_h = \left(\frac{\theta}{|\theta_g|}+1\right).$$

with $Se_{b,i} = \dfrac{k_0}{\beta}\exp\left(-\dfrac{E}{RT_{b,i}}\right)$ – Semenov number at

stationery temperature (point of ignition).

Obviously, catalytic ignition occurs with $T > T_{b,i}$ or $\theta > 0$ (increasing catalyst temperature). On the contrary,

the catalyst cools.

Then the induction period takes the following format:

$$\varkappa_{ind} = \int_{\theta_b}^{\infty} \frac{d\theta}{\left(\dfrac{1+Se_{b,i}}{1+Se_{b,i}\exp\theta}\right)\exp\theta - \left(\dfrac{\theta}{|\theta_g|}+1\right)}, \qquad (6)$$

With $\theta < 1$ the function $\bar{q}_{ch}$ may be expanded into Maclaurin's series

$$\left(\frac{1+Se_{b,i}}{1+Se_{b,i}\exp\theta}\right)\exp\theta \approx 1 + \frac{1}{1+Se_{b,i}}\theta + \frac{1-Se_{b,i}}{2(1+Se_{b,i})^2}\theta^2.$$

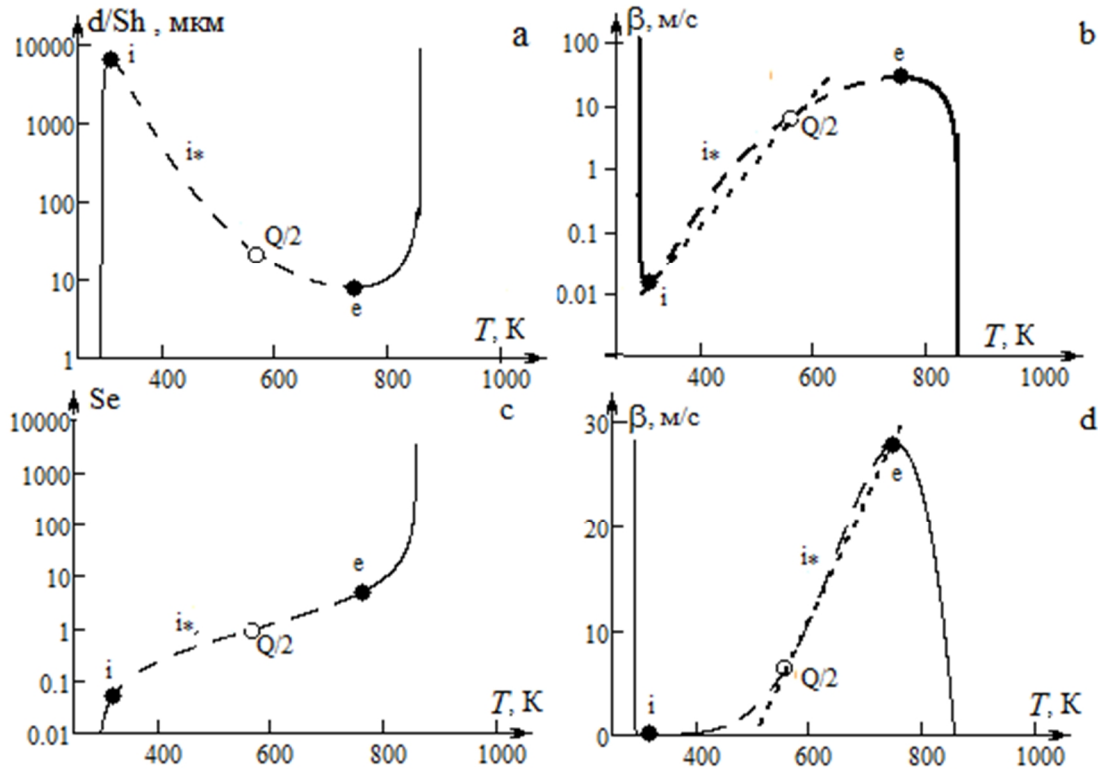So, the (6) takes the form (7), as follows,

Fig. 3. Relations between a) specified diameter, b) stationery mass exchange coefficient, c) Semenov value and d) stationery heat exchange coefficient, and platinum catalyst stationery temperature in gaseous mixture at $T_g = 293K$ with hydrogen ratio $Y_a = 0.0019$. Dashed curve: Fig.3b – (3a), Fig.3d – (3b).

$$\varkappa_{ind,1} \approx \int_{\theta_b}^{\infty} \frac{2d\theta}{\dfrac{1-Se_{b,i}}{(1+Se_{b,i})^2}\theta^2 + 2\theta\left(\dfrac{1}{1+Se_{b,i}} - \dfrac{1}{|\theta_g|}\right)} = \frac{(1+Se_{b,i})|\theta_g|}{(|\theta_g|-(1+Se_{b,i}))}\ln\left(1+\frac{2(|\theta_g|-(1+Se_{b,i}))(1+Se_{b,i})}{\theta_b|\theta_g|(1-Se_{b,i})}\right), \qquad (7)$$

The dimensional form of the above formulae is, as follows,

$$\tau_{ind,1} = \tau_{ch}\frac{(1+Se_{b,i})|\theta_g|}{(|\theta_g|-(1+Se_{b,i}))}\ln\left(1+\frac{2(|\theta_g|-(1+Se_{b,i}))(1+Se_{b,i})}{|\theta_g|\theta_b(1-Se_{b,i})}\right), \qquad (8)$$

For minor Semenov values $Se_{b,i} \ll 1$

Table 1

Catalyst ignition delay stages duration for gaseous mixture with hydrogen admixture on platinum wire
(T_b = 470 K, d/Sh = 200 mkm, T_g = 293 K)
Combustible gas admixture ratio effect

Y_a	$T_{b,i}$, K	T_m, K	θ_b	θ_g	$Se_{b,i}$	τ_1/ $\tau_{1(1)}$, s/s	τ_2/ $\tau_{2(1)}$, s/s	τ_{ind}/ $\tau_{ind(1)}$, s/s
0.0019	448	862	0.73	−5.1	0.4	0.16/0.20	0.76/0.71	0.93/0.91
0.0015	467	730	0.09	−5.28	0.72	0.56/0.47	0.69/0.68	1.24/1.16

Table 2

Catalyst ignition delay stages duration for gaseous mixture with hydrogen admixture on platinum wire
(T_b = 470 K, Y_a = 0.0019, T_g = 293 K, Sh = 0.5)
Catalyst diameter effect

d/Sh, mkm	$T_{b,i}$, K	T_m, K	θ_b	θ_g	$Se_{b,i}$	τ_1/ $\tau_{1(1)}$, s/s	τ_2/ $\tau_{2(1)}$, s/s	τ_{ind}/ $\tau_{ind(1)}$, s/s
200	448	862	0.73	−5.1	0.4	0.16/0.20	0.76/0.71	0.93/0.91
400	384	863	2.11	−4.69	0.262	0.39/0.44	3.44/3.04	3.83/3.48
1000	381	864	4.06	−4.01	0.158	2.31/1.35	21.48/19.62	23.79/20.97

$$\tau_{ind} \approx \int_{\theta_b}^{\infty} \frac{2d\theta}{\theta^2 + 2\theta\left(1 - \frac{1}{|\theta_g|}\right)} = \frac{|\theta_g|}{|\theta_g| - 1} \ln\left(1 + \frac{2(|\theta_g| - 1)}{\theta_b |\theta_g|}\right).$$

It is evident that one of the necessary conditions is $|\theta_g| > 1$.

Dependence of the catalyst temperature with time on the first stage can be approximated:

$$\tau(T) = \tau_{ch} \int_{\theta_b}^{(T-T_S)E/RT_S^2} \frac{2d\theta}{\frac{1 - Se_{b,i}}{\left(1 + Se_{b,i}\right)^2}\theta^2 + 2\theta\left(\frac{1}{1 + Se_{b,i}} - \frac{1}{|\theta_g|}\right)} \quad (9)$$

The obtained formula (7) may be applied with $Se_{b,i} < 1$, meeting a wide range of the catalyst size.

IV. The second stage of ignition period

The second stage may be characterized as the reaction's passage into deeper diffusion area and approximation with pre-given (e.g. 1%) catalyst temperature margin δ (наприклад 1%) to stationery catalytic combustion point $T_m = T_Q + T_g$ [6].

Equation to define duration of this stage may be written, as follows,

$$\frac{V_c}{S_c} c_c \rho_c \frac{\partial T}{\partial \tau} = Q_a \left(1 + \psi_T\right) k \rho_{gs} Y_a \frac{1}{Se} - \alpha\left(T - T_g\right), \quad (10)$$

Duration of this stage is calculable as an integral

$$\tau_2 = \frac{V_c c_c \rho_c}{S_c \alpha} \int_{T_2}^{T_m(1-\delta)} \frac{dT}{T_Q + T_g - T},$$

The lower border of the integral may be assigned to initial catalyst temperature without substantial error $T_2 = T_{b,i} + RT_{b,i}^2 / E$. The resulting duration of this stage may be defined, as follows:

$$\tau_2 = \frac{V_c c_c \rho_c}{S_c \lambda_g Le^m} \frac{d}{Sh} \ln\left(\left(1 - \frac{T_{b,i} + RT_{b,i}^2 / E}{T_m}\right) \cdot \frac{1}{\delta}\right), \quad (11)$$

Tables 1 and 2 compare duration of the 1st (8) and 2nd (11) stages and time of catalytic ignition in general with changes in combustible gas ratio and catalyst diameter, respectively. Satisfactory coordination is observed. Thus, point of ignition is the essential value to be identified at the earliest preliminary stage. While the duration of the 2nd stage is practically independent on it, the duration of the 1st stage is determined by difference between initial temperature and ignition point. As it is obvious from (7) and (8), the duration of the 1st stage changes rapidly with the increase of difference $T_b - T_{b,i}$.

Conclusion

This is to propose an analytical method to determine gaseous mixtures catalytic ignition time on metallic catalysts with combustible gaseous admixtures based on preliminary calculation of ignition temperature. The method is based on the preliminary finding of the critical ignition temperature. The total ignition delay time is split into two stages: catalyst preheating in transient and diffusion areas. Analytical formulae are developed for each stage duration, enabling to evaluate initial catalyst temperature, concentration admixture's combustible gas and particle (wire) diameter effects. The ignition of the catalytic reaction on the catalyst surface in cold gas mixtures by pre-heating only the catalyst is energetically advantageous than constant heating of the gas mixture in order to realize catalytic self-ignition.

Kalinchak V.V. - Doctor of Physics and Mathematics, Professor of the Department of Thermophysics;
Chernenko A.S. – PhD (physical and mathematical sciences), associate professor of thermophysics department;

Sikorskyi M.V. **-** Master of Physics Department;
Sofronkov A.N. **-** Professor, Doctor of Technical Sciences, Academician of the Academy of Sciences of
the Higher School of Ukraine, Full Member of the American Academy of Sciences in New York;
Fedorenko A.V. **-** postgraduate student.

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В.В. Калінчак[1], О.С. Черненко[1], М.В. Сікорський[1], О.Н Софронков[2], А.В. Федоренко[1]

Період індукції і температура горіння холодних газоповітряних сумішей з домішками горючого газу на платиновій частинці (нитці)

[1]Одеський національний університет імені І.І. Мечникова, вул. Дворянська, 2, м. Одеса, Україна, 65000, e-mail: chernalex@ukr.net
[2]Одесский державний екологічний університет, вул. Львівська, 15, м. Одеса, Україна, 65000, teplophys@onu.edu.ua

У роботі аналітично знаходиться період індукції і температура каталітичного горіння на металевій частинці(нитці)холодних газоповітряних сумішей з домішками горючих газів на прикладі безполум'яного горіння газоповітряних сумішей з домішками водню на платинової нитці. Стійке безполум'яне горіння спостерігається через період індукції в результаті підвищення початкової температури частинки каталізатора вище критичної температури запалювання, яка залежить від приведеного діаметра частинки і концентрації домішки горючого газу. Весь час виходу на дві частини: при протіканні каталітичної реакції в перехідній і дифузійній областях. Показано, як впливає концентрація домішки горючого газу, приведений діаметр каталізатора та перевищення початкової температури над температурою запалювання на тривалість окремих стадій та період індукції.

Ключові слова: металеві каталізатори окислення, платинова нитка, водень, період індукції, запалювання , самозаймання, погасання, поверхневе (безполум'яне) горіння.