Introduction

Multiple heterogenous chemical oxidation reactions and flameless combustion of combustible gases admixtures occur on porous catalyst particles staying within gas/air mixture flow. Critical conditions of flameless combustion depend on size of the particle specific surface area and on combustible gases concentration. Platinum-group metals and metallic oxides of variable valence act as catalysts. Platinum-group metals are applied either in the form of threads, or are speckled into carrier’s particles. The majority of active areas are located inside pores in oxidation catalyst [1]. Catalytic reaction occurs after the reagent molecules penetrate through the boundary level surrounding catalyst’s particles (external diffusion) and afterwards through the particle’s pores (internal diffusion). The resulting interaction between thermal conductivity, reagents diffusion, reaction products and chemical reaction plays an important part in catalyst’s efficiency. Ingredients transfer inside the catalyst should be taken into account if the pores’ diameter exceeds 50 nm. Such solid catalyst particles with large pores are applied, for instance, in oxidation reactions, membrane reactors and in certain biological applications.

Porosity and nanostructure are the most universal peculiarities of heterogenous solid catalysts capable to determine accessibility of particular active areas, reaction mechanism and selectivity of desirable products [2]. However, thermal radiation losses should be taken into account for large catalyst packs, even with comparatively low radiation factor typical for metallic oxidation catalysts (ε = 0.1 - 0.2).

Problem of diffusion inside porous catalyst particles is dealt with for a rather long time [3-5]. Mostly, attention is focused on simultaneous review of convection processes, diffusion processes and reaction flow inside porous catalyst. However, critical conditions of catalytic reaction’s steady running and porosity effect upon reaction characteristics are generally neglected.

This work is aimed to establish the catalyst porosity effect (mean specific surface of pores) upon crucial characteristics of high-temperature heat and mass exchange and kinetic parameters of heterogenous catalytic combustion for minor combustion gases’ quantities in heated air. Here the attention is also paid to combustible gases external and internal mass transfer to the particle surface and into the depth of chemical reaction penetration into the particle.
Research objective

The review is carried on referring to ammonia oxidation reaction on platinum catalyst:

\[ \text{NH}_3 + \text{O}_2 \xrightarrow{\text{Pt}} \text{NO} + \text{H}_2\text{O} \]

with parameters, as follows \( Q_o = 13.3 \text{ MJ/kg NH}_3, E = 139 \text{ kJ/mole}, k_o = 13 \times 10^{13} \text{ m/sec}. \)

According to contact principle, suppose that active combustible component molecules of gaseous environment collide with oxygen molecules absorbed by the catalyst surface generating reaction products of heterogenous catalytic reaction. Rate of heterogenous catalytic reaction on the catalyst particle surface and on the surface with combustible component available in gaseous environment is directly related with relative mass concentration of such a combustible component close to catalyst’s surface \( Y_{as} \):

\[ W_a = W_{as} + W_{av} = k \sum p_g s Y_{as} = \left( k + k_v \right) p_g s Y_{as} \cdot \]

with \( W_a, W_{as}, W_{av} \) – total rate of catalytic reaction for combustible substance of the outside surface and at the pores’ surface, respectively, kg/(m²·sec); \( k, k_v \) – internal reaction of combustible with oxygen total, actual and efficient constant values, respectively, m/sec; \( T \) – catalyst’s temperature, K; \( \rho_{gs} \) – gaseous mixture density near the particle’s surface kg/m³; \( P \) – pressure, Pa.

Heterogenous catalytic reaction speed constant in this case is represented by a sum of reaction constants on the external surface and inside pores.

Actual catalytic reaction speed constant on the external surface and on pores surface of catalyst’s particle increases as the temperature grows subject to Arrhenius equation:

\[ k = k_o \exp \left( -\frac{E}{RT} \right) , \]

with \( k_o \) – pre-exponential multiplier, m/sec; \( E \) – activation energy, J/mole; \( R \) – universal gas constant, J/(mole K).

Heat conductivity equation for internal reactions and reaction running on the surface of a porous catalyst particle may be represented, as below:

\[ c_r \rho_r \frac{T}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 k_c \frac{\partial T}{\partial r} \right) + \frac{q_{ch,p}}{\rho_r}, \]

\[ T(t) = T_o, \]

\[ q_{ch,p} = Q_j \frac{1}{V_j} k_{gs} s Y_a = Q_j F_i k_{gs} s Y_a, \]

with \( s \) – particle shape factor (0 – plate, 1 – cylinder, 2 – sphere), \( F_i \) – specific pores’ area (total pores’ surface area value divided by particle’s volume), m⁻¹; \( Q_j \) – reaction’s thermal effect per 1 kg of combustible gas, J/kg; \( N \) – number of pores with \( S \), surface area; \( V_c \) – catalyst’s volume, \( q_{ch,p} \) – heat generation power on pores’ surface per particle’s volumetric unit, W/m³.

General heat flow in the particle’s center takes a zero value:

\[ -r^4 \lambda_c \frac{\partial T}{\partial r} \bigg|_{r=0} = 0. \]

Boundary condition on catalyst surface may be represented, as below, taking into account reaction running on its surface:

\[ -\lambda_c \frac{\partial T}{\partial r} \bigg|_{r=r_d/2} + q_{ch,s} = q_g + q_w, \]

\[ q_w = \varepsilon \sigma \left( T^4 - T_w^4 \right), \]

\[ q_{ch,s} = Q_j k_{gs} s Y_{as}. \]

The problem may be simplified by means of taking into account correlation between heat conductivity factors for gaseous mixture and solid catalyst and between thermal conductivity factors. In case when the catalyst heat conductivity factor exceeds substantially the respective heat conductivity of gaseous mixture the temperature distribution inside the catalyst may be ignored. Thus dependence of temperature on coordinate may be ignored. It enables to average temperature throughout the entire catalyst volume. Thus, non-steady state heat balance equation for porous catalyst may be written in the format, as follows:

\[ \frac{d}{6} c_r \rho_r \frac{dT}{dt} = q_{eff}, T(t=0) = T_b. \]

\[ q_{eff} = q_{ch} - q_b, \]

\[ q_{ch} = q_{ch,s} + q_{ch,v}, \]

\[ q_{ch,v} = Q_j W_{av}, W_{av} = k_{gs} F_i \frac{d}{6} \left( Y_i \right) = k_{gs} s Y_{as}. \]

with \( q_{eff} \) – efficient volumetric heat release rate, W/m².

Internal diffusion problem should be solved to calculate internal reaction rate and combustible substance concentration profile inside the particle:

\[ \rho_r \frac{\partial Y_a}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 D_i \frac{\partial Y_a}{\partial r} \right) - W_a, \]

with existing boundary conditions inside the particle:

\[ r^4 D_i \rho_{gs} \frac{\partial Y_a}{\partial r} \bigg|_{r=0} = 0. \]

And on the particle’s surface

\[ W_{av} = D_i \rho_{gs} \frac{\partial Y_a}{\partial r} \bigg|_{r=r_d/2}, W_{as} + W_{av} = j_a, \]

\[ W_{as} = k_{gs} s Y_{as}. \]

Referring to outlined film model, combustible component mass flow density \( j_o \) on the catalyst surface (mass transferred per time unit to the catalyst’s surface...
The second expression taken in brackets may be ignored in comparison with \( 1 \). The resulting internal reactive constant increases in linear fashion as the particle diameter increases:

\[
k_i = \frac{2D_c}{d} \frac{S_e^2}{3} = k_F \frac{d}{6}. \tag{10}
\]

With \( S_e < 1 \) fault of formula (10) amounts to 6 \%, and it may be applied for \( S_e < 1.5 \), within up to 15 \% accuracy.

Further increase in temperature and particle diameter may lead to chemical reaction kinetics shifting to the internal diffusion zone, where \( S_e > 1 \). For \( S_e > 5 \) we have \( cthS_e \approx 1 \). Thus internal reactive constant in this case does not depend on the particle’s diameter:

\[
k_i = \frac{2D_c}{d} \frac{S_e^2}{3} = \frac{2D_c}{d} \frac{kF_r d^2}{4D_c} = \sqrt{kD_c F_r}. \tag{11}
\]

For \( S_e > 2 \) and \( S_e < 5 \) internal reactive constant may be described approximately with relation showing proportional relationship with the particle’s diameter:

\[
k_i = \frac{D_c}{d} \frac{S_e^2}{3} \approx \frac{kF_r d^2}{4D_c} = \sqrt{kD_c F_r}.
\]
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\[ k_v = \frac{2D_v}{d} \left( \text{Se}_v - 1 \right) = \frac{2D_v}{d} \left( \frac{kF_v d^2}{4D_v} - 1 \right) = \sqrt{kD_v F_v} - \frac{2D_v}{d} \]  
\[ \text{Thus, approximated internal reactive constant value may be applied in analytical calculations depending on Se}_v, \text{ criterion value.} \]

Stationary stable and critical states

Here all the stationary states (both stable and critical) are reviewed referring to the catalyst temperature. They are determined by combustible component concentration. In this view, relation is subject to review between combustible substance concentration and catalyst temperature obtained from stationary condition (4):

\[ Y_a = \left( \frac{\beta}{k + k_v} + 1 \right) \left( \frac{c_g}{Q_a L e^{1-m}} + \frac{v \sigma}{Q_a \rho_g} \left( T - T_g \right)^4 \right) \]  

Catalytic combustion temperature and low-temperature oxidation increase as the oxidant concentration grows Fig. 1). The time-independent relation \( Y_a(T)_i \), indicates, that maximum point corresponds to critical condition of gases self-ignition (point i), and minimum point corresponds to critical condition of self-fading (point e).

Abnormal drop in catalyst stationary temperature with increasing ammonium concentration \( Y_a \) is observable within \( Y_a < Y_a < Y_{aw} \) interval (unstable critical states corresponding to critical initial catalyst temperature \( T_{bi} \)). With catalyst initial temperature exceeding the value corresponding to stationary line \( Y_a(T_{bi}) \), the catalyst temperature grows until it achieves stable catalytic combustion temperature. If initial temperature \( T_{bi} < T_{bi} \), the catalyst temperature will decrease close to low-temperature oxidation value.

Catalyst porosity reduces critical concentration value

\[ Y_a = \left( \frac{\beta}{k + k_v} + 1 \right) \left( \frac{c_g}{Q_a L e^{1-m}} + \frac{v \sigma}{Q_a \rho_g} \left( T - T_g \right)^4 \right) \]  

Fig. 1. Relations between a) stationary temperature and b) reaction penetration depth into pores and combustible substance concentration (line 1) and ignoring the porosity (line 2); particle diameter \( d_i = 1000 \mu m \) at \( T_{gi} = 420 \text{ K}, T_{gi} = 293 \text{ K}, F_{vi} = 10^1 \text{ m}^3, \chi = 0.1 \).

Table 1

| Porosity                | Formula applicable for \( k_v \) calculation | \( T_g = 420 \text{ K}, T_w = 293 \text{ K} \) \((T_i = 426 \text{ K})\) | \( T_g = 400 \text{ K}, T_w = 293 \text{ K} \) \((T_i = 406 \text{ K})\) |
|-------------------------|---------------------------------------------|----------------------------------------------------------------|----------------------------------------------------------------|
| Solid particle \( F_{vi} = 0 \) | –                                          | 3.65                                                           | –                                                               |
|                        | (8)                                        | 2.86                                                           | 16.5                                                            |
| Porous particle \( F_{vi} = 10^3 \text{ m}^3 \) | (10)                                      | 2.76                                                           | 16.4                                                            |
|                        | (11)                                       | 2.34                                                           | 8.87                                                            |
|                        | (12)                                       | 2.98                                                           | –                                                               |
| Porous particle \( F_{vi} = 10^4 \text{ m}^3 \) | (8)                                        | 1.55                                                           | 6.00                                                            |
|                        | (10)                                       | 0.92                                                           | 5.11                                                            |
|                        | (11)                                       | 1.42                                                           | 4.12                                                            |
|                        | (12)                                       | 1.55                                                           | 6.31                                                            |

\( Se_v \approx 0.6 \) by (9) \( Se_v \approx 2.2 \) by (9)
required for ammonium ignition in air on the catalyst particle. In this case chemical reaction in low-temperature zone runs practically within entire catalyst volume up to the self-ignition point. It is illustrated by $Se$ proximity to 1 (Table 1, Fig. 1b). Fading concentration critical value does not practically depend on the catalyst porosity.

It is rather difficult to apply formula (8) to analytical search for critical states. Referring to Table 1 approximated relations (10), (11) and (12) may be applied to identify critical states depending on particle specific porosity surface area and surrounding gaseous mixture temperatures. Formula (10) with $Se_{vi} < 2$ may be reasonably applied to calculate internal reactive constant. Formula (11) is applicable for $Se_{vi} > 2$. As the gaseous mixture temperature decreases, even by 20 K only, the internal Semyonov value number for critical point decreases triple.

Fig. 2 represents critical states for ammonium ignition in air on catalyst particle depending on the particle dimensions with various porosity, as a plurality of points being extremum of function (13). Taking into note thermal losses for radiation leads to formation of diameter limiting value for gases self-ignition zone on catalyst particles surface. Applying porous catalyst, even with small specific pores surface area (e.g., $10^3 \text{ m}^{-1}$) leads to substantial decrease of minimum critical value of combustible component concentration, which, being exceeded, provokes catalytic ignition on the catalyst surface. Particle diameter increases with porosity increase.

It should be noted, that critical states for self-ignition correspond to external kinetic area where the chemical reaction runs and to internal diffusion area $(d_{vi} > 300 \mu\text{m}, Se_{vi} > 2)$. It enables to apply formula (11) to identify location of minimum point to calculate internal reactive constant.

Stationary condition taking into account porosity looks, as below:

$$Q \frac{Y_{av}}{\rho_y} \left( k + \sqrt{kD_v F_v \frac{2D_v}{d}} \right) = \alpha(T - T_g) - \varepsilon\sigma(T^4 - T_w^4).$$

(14)

Alteration made in a formula part indicating porosity of an item:

Fig. 2. Relations between a) combustible substance critical concentration and b) catalyst critical temperature, c) internal Semyonov value number and d) Semyonov value number and diameter at $T_g = 420 \text{ K}, T_w = 293 \text{ K}$. Curves: 1 – solid particle; 2 – porous particles, $F_v = 10^3 \text{ m}^{-1}, \chi = 0.1$; and 3 – $F_v = 10^4 \text{ m}^{-1}, \chi = 0.1$. 

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\[ \frac{2D}{d} = b\alpha, \text{ where } b = \frac{D}{3\lambda g \text{Nu}}, \quad \alpha = \beta c g \rho g \text{Le}^{1-m}. \]

Thus stationary condition may be described, as follows:

\[ Q_a Y_{a\bar{\rho}} \left( k + \sqrt{kD_F V - b\alpha} \right) = \alpha \left( T - T_s \right) + \varepsilon \sigma \left( T^4 - T_s^4 \right). \]

The equation \( \frac{\partial q_{ch}}{\partial \alpha} = \frac{\partial q_b}{\partial \alpha} \) may be applied to identify minimum critical state point, with \( q_{ch} \) and \( q_b \) - left and right parts of the equation (14). Thus,

\[ -Q_a Y_{a\bar{\rho}} b = \left( T - T_s \right). \]

Temperature corresponding to minimum point (minimum ignition point \( t_{im} \)): \( T_{im} = T_s - \frac{Q_a Y_{a\bar{\rho}} D}{3\lambda g \text{Nu}}. \)

\[ Y_{a_{im}} = \frac{\varepsilon \sigma \left( T_s^4 - T_{im}^4 \right)}{Q_a Y_{a\bar{\rho}} k_0 \left( 1 + \frac{D_F}{\sqrt{k(T_s)} d \cdot k(T_s)} - \frac{2D}{d} \right)} - 1 \exp \left( \frac{E}{RT_s} \right). \] \hspace{1cm} (15)

\[ \frac{\partial q_{ch}}{\partial T} = \frac{\partial q_b}{\partial T} \] may be applied to calculate critical diameter in the minimum point.

\[ Q_a Y_{a\bar{\rho}} \left( k + \frac{1}{2} \sqrt{kD_F V} \right) \cdot \frac{E}{RT_s^2} = \alpha + 4\varepsilon \sigma T^3. \]

Applying \( T_{im} \approx T_s \) and (13), diameter may be calculated, as follows:

\[ \frac{\varepsilon \sigma \left( T_s^4 - T_{im}^4 \right)}{k} \left( \frac{k + \frac{1}{2} \sqrt{kD_F V}}{k - \sqrt{kD_F V} - b\alpha} \right) \left( \frac{E}{RT_s^2} \right) = \alpha \frac{RT_s^2}{E} + 4\varepsilon \sigma T^3 \frac{RT_s^2}{E}. \]

Applying (16), diameter is given by:

\[ \frac{1}{d} \left( \frac{\lambda g \text{NuRT}_s^2}{E} \varepsilon \sigma T_s^3 \right) = \left( 1 - \frac{T_{im}^4}{T_s^4} \right) \left( \frac{k(T_s) + \frac{1}{2} \sqrt{k(T_s)D_F V}}{k(T_s) + \sqrt{k(T_s)D_F V} - 2D_k} \right) - 4RT_s \frac{RT_s^2}{E}. \] \hspace{1cm} (16)

Table 2 contains comparative combustible concentration values and catalyst particle diameter describing border conditions of ignition calculated sing formulae (15) and (16).

Comparing the obtained formulae with similar in which reaction in pores was not taken into account \[8\], enables to identify a multiplier defining porosity effect on mixture concentration limit value and catalyst particle diameter:

\[ Y_{a_{im}} = \left( 1 + \frac{D_F}{\sqrt{k(T_s)} d \cdot k(T_s)} - \frac{2D}{d} \right)^{-1}. \]

\[ \frac{1}{d_{im}} = \left( \frac{k(T_s) + \frac{1}{2} \sqrt{k(T_s)D_F V}}{k(T_s) + \sqrt{k(T_s)D_F V} - 2D_k} \right). \] \hspace{1cm} (17)

So, as the pores specific surface area increases value \( Y_{a_{im}} \) decreases. Catalyst material porosity effect \( \chi \) according to (17) is more complicated.

**Catalyst porosity effect upon induction period**

Fig. 3 illustrates time relation of catalyst temperature \( T \) and Semyonov value \( \bar{S}\bar{e} \) during ammonium catalytic oxidation both taking into account and ignoring particles porosity for catalytic ignition case. Apparently, porous particle naturally reduces time of high-temperature oxidation mode gaining (induction period).

Total time period may be split into three stages. The first represents inert preheating. Semyonov value is extremely small indicating minor chemical heat emission ratio. The second stage represents particle’s chemical heating due to chemical heat emission as chemical
reaction runs in kinetic zone. The third stage represents particle heating due to chemical heat emission as the chemical reaction runs in diffusion zone.

Porosity effect is shown in Table 3. Decrease of the first stage with increasing porosity is caused by decrease of the upper limit of this stage. Chemical reaction commences earlier due to porosity than with solid particles.

Decrease of the second stage is caused by substantial role played by internal reaction running in kinetic zone. Duration of the third stage does not depend on the particle’s porosity. Combustible component does not reach full depth of a particle with chemical reaction running in external diffusion zone.

**Conclusions**

Combustible gas oxidation reaction inside porous particle of catalyst leads to decrease in combustible gas minimum concentration, which, once exceeded provokes catalytic ignition on catalyst particle surface. The obtained formula enables to analyze porosity effect on critical value and corresponding particle diameter. Increase in catalyst porosity leads to drop of induction period due to reduction in preheating time and chemical heating time with the reaction running in kinetic zone.

| Catalyst particle porosity effect on limiting critical conditions of ammonia ignition in air \(T_g = 420\) K |
|---------------------------------|---------------------------------|
| \(F_v = 10^3\) m\(^{-1}\) | \(F_v = 10^4\) m\(^{-1}\) |
| \(Se_{v_{c}} = 10.9\), 3 Fig. 2 | \(Se_{v_{c}} = 15.1\), 3 Fig. 2 |
| Value ref. Fig. 2, solution (15), (16) | Value ref. Fig. 2, solution (15), (16) |
| \(Y_{v_{c}}\) | 0.0199 | 0.0243 | 0.0104 | 0.0114 |
| \(d_{v_{c}}\), \(\mu m\) | 2950 | 2320 | 3350 | 3350 |

**Fig. 3.** Time relations a) platinum particle temperature and b) Semyonov value with ammonium concentration \(Y_a = 6.0\%\) for \(T_g = 420\) K, \(T_c = 293\) K, \(T_b = 293\) K, \(d/\text{Nu} = 1000\) \(\mu m\). 1 – solid particle, 2 – porous particle \(F_v = 10^3\) m\(^{-1}\), \(\chi = 0.1\).

**Table 3**

| \(Y_a = 6.0\%\), \(T_g = 420\) K, \(T_b = 293\) K, \(d/\text{Nu} = 1000\) \(\mu m\), \(\chi = 0.1\) |
|--------------------------------|--------------------------------|
| \(F_v, \text{m}^{-1}\) | \(t_1, \text{sec}\) | \(t_2, \text{sec}\) | \(t_3, \text{sec}\) | \(t_{ind}, \text{sec}\) | \(T_1, \text{K}\) | \(T_3, \text{K}\) |
| 0 | 111.6 | 59.1 | 58.8 | 229.5 | 420 | 965 |
| \(10^3\) | 75.9 | 38.1 | 58.8 | 172.8 | 415 | 965 |
| \(10^4\) | 53.7 | 29.1 | 58.8 | 141.6 | 407 | 965 |
| \(10^5\) | 34.8 | 24.0 | 58.8 | 117.6 | 388 | 965 |
| \(10^6\) | 22.5 | 19.8 | 59.1 | 101.4 | 367 | 965 |

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Вплив поруватості частинки каталізатора на характеристики безполуменевого горіння

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Розглядається задача тепломасообміну поруватої частинки каталізатора, на поверхні якої, включаючи внутрішню поверхню пор, протікає необернена гетерогенна реакція першого порядку. Аналітично показано, що в нагрітій газовій суміші поруватість каталізатора сприяє зменшенню мінімальної концентрації каталітичного самозаймання домішки в суміші та збільшенням відповідного діаметру частинки каталізатора. Цій концентрації відповідає зовнішній кінетичний та внутрішній дифузійний режими протікання реакції.

Ключові слова: металеві каталізатори окислення, поруватість, займання, поверхневе (безполум'яне) горіння.