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The Effect of Operational Parameters on the Catalytic Combustion of *n*-Butane/Air Mixtures on Platinum Wire

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Abstract The effect of mixture composition, total pressure and catalyst temperature on the kinetics of the catalytic combustion of *n*-butane/air or *n*-butane/oxygen mixtures on isothermally heated platinum wire is reported. The catalyst temperature was changed from 680 to 1,130 K for mixtures containing *n*-butane between 1 and 4.5% at total pressures between 10 and 100 kPa. The measurements allowed the determination of different kinetic properties like reaction rate, turnover frequency, overall and partial reaction orders and activation energy. The pressure dependence of the reaction rate offered the possibility to make a critical analysis of the kinetic equations frequently utilized in the literature.

Keywords Catalytic combustion · *n*-Butane/air mixtures · Operational parameters · Kinetics

1 Introduction

During the last decades the catalytic combustion of higher alkanes and, in particular, of *n*-butane became an intensively studied subject, generated by promising applications [1, 2] and by necessity to extend and validate the kinetic modeling elaborated for lower alkanes [3, 4]. The lower reaction temperatures resulting in reduced NO_x emissions, improved combustion stability for lower fuel content, as well as the high and adjustable reaction rates which provide efficient operation of the practical devices are obvious benefits compared to classical gas phase combustion. The overall reaction rate of the catalytic combustion is an intricate function of many operational variables like fuel type, mixture composition, catalyst temperature, total pressure, gas phase motion etc. The aim of the present paper is to investigate the effect of fuel concentration, total pressure and catalyst temperature on the kinetics of the catalytic combustion of different stagnant *n*-butane/air mixtures on a thin platinum wire maintained in isothermal regime at temperatures significantly higher than the ignition temperatures [5, 6]. It represents an extension of a previous work dealing with the catalytic combustion of the stoichiometric *n*-butane/air mixture in a similar environment [6]. The key experimental variable measured during the combustion process is the reaction heat flow rate which is proportional to the reaction rate. The technique has the advantage of performing direct measurements of the catalytic reaction rate in strictly controlled isothermal conditions at different total pressures allowing a critical analysis of suitability of the most frequently proposed kinetic equations which consider the combustion process as a single reaction.

2 Experimental Method

The details regarding the experimental set-up referring to the reaction heat flow rate measurements on isothermally heated platinum wires and to the sample preparation were given elsewhere [5, 6]. Briefly, samples of air or fuel/air mixture are sequentially introduced into the evacuated test cell at the same initial pressures. A platinum wire, located

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in the center of the cell, is rapidly heated following a quasi-rectangular profile using the discharge of a condenser. It is subsequently maintained at a constant temperature using a power supply regulated by a feedback loop designed to maintain a constant resistance of the platinum wire. The wire with resistance R_w is connected in series with a standard resistor R_std which allows the monitoring of the input power on the wire during the isothermal heating by measuring the voltage drop U_std across it. These two resistances, coupled with a potentiometer *P* form a Wheatstone bridge maintained at equilibrium. The measurement of the reaction heat flow rate requires the recording of two U_std–time diagrams: one in mixture and one in air, at the same pressure and temperature. The recording in air is used to eliminate the contribution of heat losses through conduction and convection. The simplified outline of the experimental set-up is illustrated in Fig. 1a.

The reaction heat flow rate $\mathrm{d}Q_\mathrm{r}/\mathrm{dt}$ can be calculated as:

$$\mathrm{d}Q_\mathrm{r}/\mathrm{dt} = \left(R_\mathrm{w}/R^2_\mathrm{std}\right)\left[\left(U^2_\mathrm{std}\right)_\mathrm{air} - \left(U^2_\mathrm{std}\right)_\mathrm{mixture}\right] \qquad (1)$$

where Q_r is the heat delivered by the exothermal catalytic reaction and is related to the reaction rate r_R through:

$$r_\mathrm{R} = \frac{1}{\Delta^c H^0_\mathrm{T} \cdot S_\mathrm{w}} \frac{\mathrm{d}Q_\mathrm{r}}{\mathrm{dt}} \qquad (2)$$

where $\Delta^c H^0_\mathrm{T}$ is the standard heat of combustion and S_w the wire surface. It is obvious that $\Delta^c H^0_\mathrm{T}$ has the same value only for lean to stoichiometric mixtures.

All involved quantities are experimentally measurable. The necessary gas pressure and wire temperature are measured according to described procedures. An illustrative example with $\mathrm{d}Q_\mathrm{r}$ versus time is given in Fig. 1b. The experimental results consist in recording the $\mathrm{d}Q_\mathrm{r}/\mathrm{dt}$ versus time diagrams for *n*-butane/air or *n*-butane/oxygen mixtures at different total pressures p_0 between 10 and 100 kPa and wire temperatures T_w between 680 and 1,130 K. It should be noted that even the lowest working temperature (680 K) is much higher than the reported ignition temperatures of *n*-butane/air mixtures, between 450 and 550 K [7–9]. The samples contained 1.0, 1.5, 2.0, 2.5, 3.13, 4.0 or 4.5% *n*-butane (in volume percents) representing lean, stoichiometric (3.13%) and rich mixtures. The reaction heat flow rate $\mathrm{d}Q_\mathrm{r}/\mathrm{dt}$ presents a short continuously increasing transient period followed by a steady state value. All the data used in this paper refer to the steady state regime.

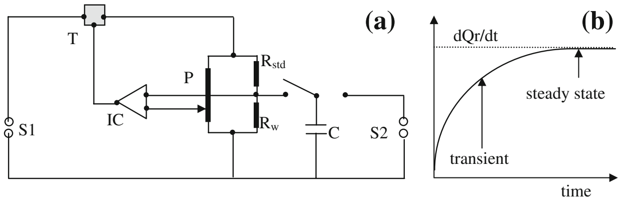

Fig. 1 **a** Schematic of the experimental set-up: S_1, main power supply; *T*, series transistor; *IC*, integrated circuit whose output adjusts the applied voltage across R_std and R_w to maintain the bridge at equilibrium; *C*, condenser for initial wire heating; S_2, secondary adjustable power supply for charging the condenser C; **b** Typical variation of reaction heat flow rate $\mathrm{d}Q_\mathrm{r}/\mathrm{dt}$

The measurement of the heat release rate for the catalytic combustion of alkanes (including *n*-butane) on platinum was often performed in order to determine the ignition temperatures [7–9]. It was assumed [6, 7] or proved [8] that for the catalytic combustion of lean to stoichiometric alkane/air mixtures on platinum, carbon dioxide and water were the only reaction products formed. Moreover, the results of several kinetic studies using the mass spectrometry to analyze the composition of the reaction mixture [10, 11] in similar conditions confirmed this stoichiometry. All these results support our hypothesis that the heat release rate is a proper measure of the catalytic combustion rate.

3 Results and Discussion

It can be seen that the increase of the reaction rate continues even for rich mixtures, although the overall stoichiometry changes. As a first approximation the reaction rate can be analyzed on the basis of the empirical power law:

$$\begin{aligned} r_\mathrm{R} &= A(p_\mathrm{F}/p^*)^{n\mathrm{F}} \cdot (p_\mathrm{ox}/p^*)^{n_\mathrm{ox}} \cdot \mathrm{e}^{-E_a/RT_\mathrm{w}} \\ &= A_0(p_0/p^*)^n \cdot \mathrm{e}^{-E_\mathrm{a}/RT_\mathrm{w}} \end{aligned} \qquad (3)$$

where *A* and A_0 are pre-exponential factors, E_a is the overall activation energy, p_F, p_ox, p^* and p_0 are the partial pressures of fuel and oxygen, the reference pressure (taken as unity here) and the total pressure, respectively, and *R* is the molar ideal gas constant. The partial pressures are given by $p_\mathrm{j} = X_\mathrm{j} p_0$ with X_j the molar fractions and the overall reaction order is approximated by $n = n_\mathrm{F} + n_\mathrm{ox}$. The overall reaction order *n* is obtained from the linear regression $\mathrm{Ln}(r_\mathrm{R})$ or Ln $(\mathrm{d}Q_\mathrm{r}/\mathrm{dt})$ against $\mathrm{Ln}(p_0/p^*)$ as will be discussed below, in connection with the dependence of r_R on the total pressure.

The partial reaction order with respect to *n*-butane, $n \approx 1$, was obtained from the linear regression $\mathrm{Ln}(\mathrm{d}Q_\mathrm{r}/\mathrm{dt})$ versus $\mathrm{Ln}(X_\mathrm{butane})$ on the data contained in Fig. 2. It is in good agreement with other results, obtained using different techniques [10, 11]. The partial reaction order with respect to oxygen cannot be evaluated from the same data due to the small relative variations of X_ox when X_butane changes from 0.01 to 0.045. In order to evaluate n_ox, a series of measurements were performed in air and in oxygen for the same *n*-butane content at the same temperature and different total pressures. A representative example is given it Table 1.

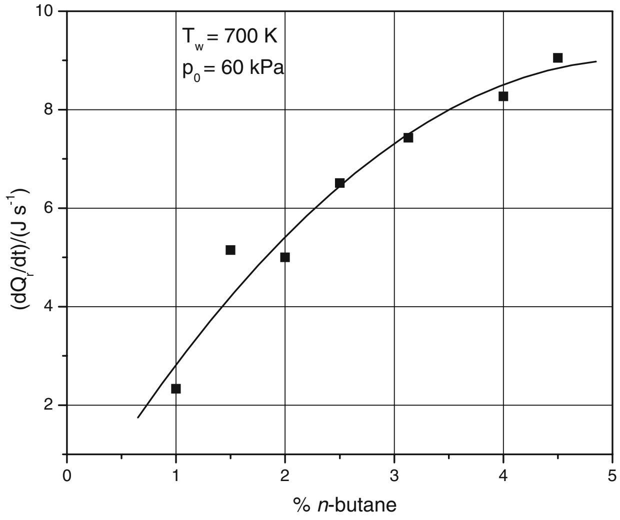

Fig. 2 The variation of the reaction heat flow rate for *n*-butane/air mixtures with the fuel content

Similar results were also obtained at lower and higher temperatures for different *n*-butane contents. It is apparent that the increase of the oxygen content has an inhibitory effect on the reaction rate, in accord with other observations [10, 11].

The partial reaction order with respect to oxygen was evaluated when X_{butane}, total pressure p_0 and wire temperature T_w were kept constants, according to:

$$dQ_r/dt = A_{0,T}(p_F/p^*)^{nF}(p_{ox}/p^*)^{n_{ox}} = aX_{ox}^{n_{ox}} \tag{4}$$

and:

$$n_{ox} = \frac{\text{Ln}\left[(dQ_r/dt)_{air}/(dQ_r/dt)_{ox}\right]}{\text{Ln}\left[(X_{ox})_{air}/(X_{ox})_{ox}\right]} = -\frac{\text{Ln}\left[(dQ_r/dt)_{air}/(dQ_r/dt)_{ox}\right]}{1.56} \tag{4'}$$

In order to match the same pressures and temperatures for both *n*-butane/air and *n*-butane/oxygen mixtures some values of dQ_r/dt were interpolated. The average value, $n_{ox} \approx -0.16$, agrees well with some previously reported data [10]. However, it should be specified that some other reported values are significantly different [6, 11], an observation commonly found for other alkanes [4]. The scatter of the reported data originates primarily in experimental conditions like composition, total pressure, catalyst temperature, which cannot be properly taken into account by an empirical kinetic equation of the form (3), usually valid only for narrow ranges of variables.

The results reflect the complexity of the overall catalytic combustion process and suggest further research to find out alternative simple kinetic equations able to explain the experimental findings. Effect of the total pressure on the reaction heat flow rates is illustrated in Fig. 3.

From the linear regression Ln(dQ_r/dt) versus Ln(p_0), at constant molar fractions of components, one obtains the overall reaction order $n = 0.25$, according to Eq. (3). For all investigated mixtures, the overall reaction order varies between 0.20 and 0.42 for different compositions and temperatures, without any apparent trend. The results will be given below in a synoptic table. The same results were also analyzed in terms of a mechanistic Langmuir–Hinshelwood equation for a competitive, non-dissociative adsorption of both reactants (each reactant molecule occupies one site), without diffusion limitations [9]:

$$r_R = k_R \frac{K_F \cdot K_{ox} \cdot p_F \cdot p_{ox}}{\cdot}(1 + K_F \cdot p_F + k_{ox} \cdot p_{OX})^2 = \frac{a_0 \cdot p_0^2}{(1 + a_1 \cdot p_0)^2} \tag{5}$$

where k_R is the rate constant of the surface reaction, K_F, K_{ox} are the adsorption–desorption equilibrium constants of fuel and O$_2$, respectively, and a_0 and a_1 are constants with obvious significances for a certain mixture at constant

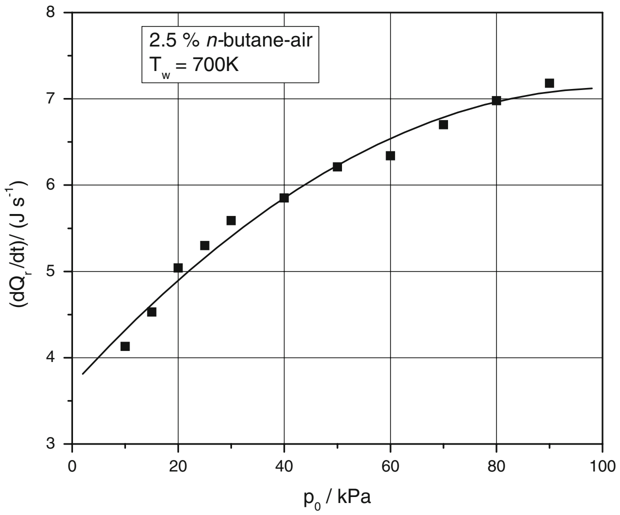

Fig. 3 Variation of the reaction heat flow rate with the total pressure

Table 1 Reaction heat flow rates at different total pressures for 2.5% *n*-butane/air and *n*-butane/oxygen mixtures at T_w = 700 K

p_0 (kPa)		10	15	20	25	30	40	50	60	70	80	90
(dQ_r/dt)/(J/s)	Air	4.13	4.53	5.04	5.30	5.59	5.85	6.21	6.34	6.70	6.98	7.18
	O$_2$	2.90	3.51	4.01	4.08	4.36	4.54	4.94	4.75	5.29	5.69	6.18
$-n_{ox}$		0.23	0.16	0.15	0.17	0.16	0.16	0.15	0.19	0.15	0.13	0.10

temperature. If $K_F p_{ox} \ll K_{ox} p_{ox}$ for alkanes [9], the significance of a_1 becomes simpler.

The assumption $K_{ox} p_{ox} \ll 1 + K_F p_F$ [9] for alkanes indicates that the reaction rate should be independent on the total pressure, in contradiction with our present results. On the other hand, if the oxygen adsorption is supposed to be dissociative (each oxygen atom occupies one site), the following kinetic equation is obtained [4]:

$$r_R = k_R \frac{k_F \cdot p_F \cdot \sqrt{K_0 p_{ox}}}{\left(1 + K_F \cdot p_F + \sqrt{K_0 \cdot p_{ox}}\right)^2} = \frac{a_0 \cdot p_0^{3/2}}{\left(1 + a_1 \cdot p_0 + a_2 \cdot p_0^{1/2}\right)^2} \quad (6)$$

where K_O is the dissociative adsorption–desorption equilibrium constant of oxygen. Assuming some further simplifications within the steady state approximations [12], simpler kinetic equations can be obtained and confronted with experimental data. Equations (5) and (6) have been more frequently utilized to describe the kinetics of the catalytic combustion of alkanes [4, 9]. The availability of the method presented above to measure, moderately easily, the reaction heat flow rate at different total pressures offers the possibility to discriminate between the above models. The reaction heat flow rate dQ_r/dt was measured for several fuel/air mixtures containing 1.5, 2.5, 3.0 and 3.12% *n*-butane at pressures between 10 and 90 kPa and at temperatures between 600 and 900 K. Three kinetic equations, representing the power law (PL): dQ_r/dt $= a_0(p_0)^{a_1}$, the Langmuir–Hinshelwood law for non-dissociative adsorption (LH1L): $dQ_r/dt = \frac{a_0 \cdot p_0^2}{(1+a_1 \cdot p_o)^2}$ and the Langmuir–Hinshelwood law for dissociative adsorption of oxygen (LH2L): $dQ_r/dt = \frac{a_0 \cdot p_0^{3/2}}{\left(1+a_1 \cdot p_0 + a_2 \cdot p_0^{1/2}\right)^2}$ were fitted on the experimental data. A representative result is given in Table 2.

The best statistical data (highest coefficient of determination, r^2, lowest fit standard error and highest $F_{\text{statistic}}$) are obtained for the empirical power law. If the two mechanistic Langmuir–Hinshelwood laws are compared to each other, the law for dissociative adsorption of oxygen (LH2L) apparently seems to fit better the experimental data, using three adjustable parameters. However, the corresponding fitting parameters take unrealistic high values and are also associated with still larger errors. Discrimination on the basis of Akaike's information criterion [13] indicates that the model LH2L is fitted significantly better by the experimental data given in Table 2. However, only ~50% of the experimental data follow the same trend, while the other half indicates the model LH1L as the most probable. It can be concluded that the mechanistic Langmuir–Hinshelwood models offer a satisfactory explanation of the experimental data without the possibility to discriminate between them. These limitations arise primarily from the simplifying assumptions, like the existence of quasi-equilibrated steps, rate determining step, the most abundant reaction intermediate, implied in deriving the kinetic equations. The usefulness of the mechanistic Langmuir–Hinshelwood models results also from the analysis of dQ_r/dt dependence on the molar fraction of *n*-butane in air at constant pressure. An example is given in Table 3.

The Langmuir–Hinshelwood equation for a competitive, non-dissociative adsorption of both reactants, $dQ_r/dt = \frac{a_0 \cdot X_F \cdot (1-X_F)}{(1+a_1 \cdot X_F)^2}$, with X_F as *n*-butane molar fraction, was fitted on the data and the following results were found: $a_0 = 286 \pm 30$, $a_1 = 3.4 \pm 1.7$, $r^2 = 0.971$; $F_{\text{statistic}} = 170$, indicating a good correlation between variables.

Taking into account the large dispersion of the data accompanying the measurements in this type of experiments, no further processing of the results was attempted.

The discrimination between different kinetic models can be accounted for using an extension of the analysis of the chemical heat flow rate versus time diagram, given in Fig. 1b and rationalized in the previous paper [6].

Assuming that the change of the surface concentration of the fuel, $C^*_{F,S}$ is given by the difference between the diffusion rate, r_D, and the catalytic reaction rate, r_s, one obtains [6]:

Table 3 Variation of dQ_r/dt with *n*-butane molar fraction in air at $T_w = 700$ K and $p_0 = 60$ kPa

X_{butane}	0.010	0.015	0.020	0.025	0.031	0.040	0.045
dQr/dt (J/s)	2.33	3.15	5.00	6.51	7.43	8.27	9.05

Table 2 Fitting parameters and statistical data for the nonlinear regression analysis of a 2.5% *n*-butane/air mixture at 700 K for pressures between 10 and 90 kPa

Model	a_0	a_1	a_2	r^2	Fit standard error	$F_{\text{statistic}}$
PL	2.39 ± 0.06	0.243 ± 0.007	–	0.994	0.081	1,492
LH1L	0.436 ± 0.082	0.242 ± 0.025	–	0.943	0.249	150
LH2L	1.43e16 ± 9.98e21	5.14e06 ± 1.80e12	9.03e07 ± 3.16e13	0.986	0.129	290

$$\frac{dC_{F,S}}{dt} = \phi \cdot \frac{dC^*_{F,V}}{dt} = k_D\left(C^0_{F,V} - C^*_{F,V}\right) - k_r \cdot C^*_{F,V} \tag{7}$$

where $C^*_{F,V}$ is the volume fuel concentration within the layer adjacent to catalyst surface, $C^0_{F,S}$ is the corresponding bulk concentration, k_D is the mass transfer constant, k_r is an overall catalytic constant of a first order reaction and ϕ is a characteristic length required to define a gas volume numerically equal with the catalyst surface. Integration of this equation followed by the truncation for the steady state regime yields [6]:

$$\frac{dQ_r}{dt} = a + \frac{\Delta^c H^0_T \cdot S \cdot k_D \cdot k_r \cdot C^0_{F,V}}{k_D + k_r} \tag{8}$$

where a is an additional small term contributed by the combustion during the temperature jump from T_0 to T_w. This equation is in agreement with the first order found with respect with n-butane but ignores the contribution of oxygen concentration. To account also for this effect, the catalytic reaction rate can be generalized as:

$$r_R = k^* \cdot C^*_{F,V} \tag{9}$$

where $k^* = k_r$ for a first order kinetics as before, $k^* = k_r \cdot C^m_{ox,V}$ for a power law equation, with m being the partial reaction order with respect to oxygen, or $k^* = k_r \cdot C^0_{ox,V} \Big/ \left(1 + K_{ox} \cdot C^0_{ox,V}\right)^2$, or other related forms for Langmuir-Hinshelwood models.

For excess oxygen, when $C^0_{ox,V} \approx$ const, the similar integration gives:

$$\frac{dQ_r}{dt} = a + \frac{\Delta^c H^0_T \cdot S \cdot k_D \cdot k^* \cdot C^0_{F,V}}{k_D + k^*} \tag{10}$$

characteristic for a mixed controlled process.

For diffusion and kinetically controlled processes, ($k_D \ll k^*$), ($k_D \gg k^*$), respectively, the reaction heat flow rate will be given by:

$$\frac{dQ_r}{dt} = a + \Delta^c H^0_T \cdot S \cdot k_D \cdot C^0_{F,V}$$

and

$$\frac{dQ_r}{dt} = a + \Delta^c H^0_T \cdot S \cdot K^* \cdot C^0_{F,V} \tag{11}$$

Converting the bulk molar concentrations in partial pressures, $p^0_F = X^0_F \cdot p_0$, $p^0_{ox} = X^0_{ox} \cdot p_0$ one obtains that for a diffusion controlled process the reaction rate should be of first order with respect to total pressure at constant X_F, $\frac{dQ_r}{dt} = a + b \cdot p_0$, irrespective of the form adopted for k*. This conclusion is in disagreement with our experimental results ($\frac{dQ_r}{dt} = a + b \cdot p^n_0$, with $0 < n < 1$). A similar dependence of the reaction rate was reported for methane catalytic combustion [14]. On the other hand a kinetically controlled process is not consistent with the high values of the reported reaction rates. A mixed control, when both k_D and k^* in Eq. (10) have significant contributions, seems to describe better the experimental findings although the explicit forms of Eq. (10) are different from Langmuir–Hinshelwood kinetic equations. This could explain the poor fitting of these equations on the experimental data as compared to the power law, known as a flexible function within a narrow range of variables. It should be finally emphasized that all these difficulties to find an adequate kinetic equation originates in the attempt to describe the result of a large number of steps using only few overall steps. Effect of the catalyst temperature is illustrated in (Fig. 4).

From the regression Ln(dQ_r/dt) versus 1/T_w, at constant pressure, according to $dQ_r/dt = A_0 \cdot (p_0/p^*)^n \cdot e^{-E_a/RT_w}$, one obtains $E_a = 6.84 \pm 0.37$ kJ/mol, characteristic for a diffusion controlled process [6].

The overall results are given in Table 4 together with the stoichiometric ratio, ϕ and the turnover frequency *TOF*. This was evaluated as $TOF = r_R/\Gamma_s$, at $T_w = 700$ K and $p_0 = 60$ kPa. The reaction rate r_R was calculated according to Eq. (2) with $S_w = 1.41 \cdot 10^{-5}$ m^2 and $\Delta^c H^0 = 2{,}658.5$ kJ/mol [6]. The surface density of the adsorption centers was taken as $\Gamma_s = 2.49 \cdot 10^{-5}$ mol/m^2 [10], neglecting the platinum surface roughening during the catalytic combustion [6]. The values of *TOF* indicate the high efficiency of platinum as catalyst for n-butane combustion. If the reaction rates r_R are compared to the fuel collision frequency with the platinum surface covered preponderantly with oxygen, calculated according to $\nu_F = p_F/(2\pi M_F R T_w)^{1/2}$ with M_F the molar mass of the fuel, it is found that r_R/ν_F takes values between 0.004 and 0.007, much lower than the reported sticking coefficients [4], indicating that some other factors contribute to this result.

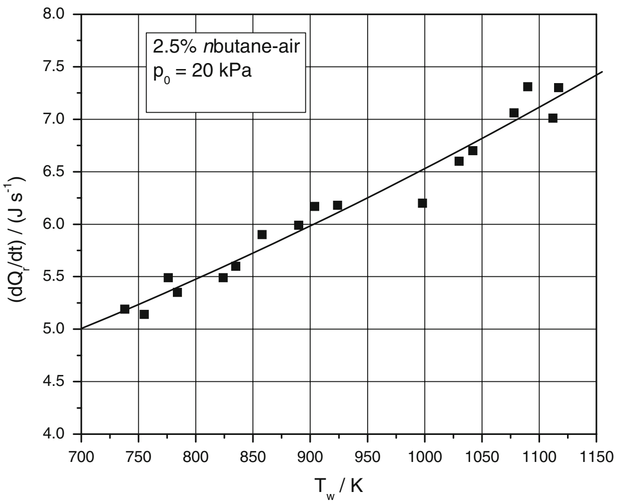

Fig. 4 Variation of the reaction heat flow rate with the catalyst temperature

Table 4 Kinetic parameters for the catalytic combustion of *n*-butane/air mixtures

% *n*-butane	ϕ	n (T_w)	E_a (kJ mol^{-1})	R_R (mol m^{-2} s^{-1})	ν_F (mol m^{-2} s^{-1})	10^{-3} *TOF* (s^{-1})
1.0	0.310	0.20 ± 0.02 (940 K)	6.17 ± 1.39	0.062	13.03	2.49
1.5	0.471	0.29 ± 0.01 (910 K)	6.40 ± 0.47	0.137	19.55	5.50
2.0	0.632	0.30 ± 0.07 (874 K)	8.80 ± 1.58	0.133	26.06	5.34
2.5	0.794	0.30 ± 0.05 (924 K)	6.84 ± 0.37	0.174	32.58	6.60
3.13	1.000	0.36 ± 0.02 (880 K)	8.17 ± 0.20	0.198	40.79	7.95
4.0	1.289	0.37 ± 0.04 (720 K)	8.43 ± 1.03	0.221	52.13	8.86
4.5	1.458	0.42 ± 0.04 (760 K)	–	0.241	58.64	9.68

Within the limits of experimental errors there seems to be no significant trend in the variations of the overall parameters with *n*-butane concentration. The activation energies are all within the range of diffusion controlled processes, as expected for these high temperatures (680–1,130 K), while the overall reaction order indicates that the reaction rate increases significantly with the total pressure.

As a general remark it should be specified that the discussion refers specifically to lean and stoichiometric mixtures and was extended also to two rich mixtures, although these have a different stoichiometry, to observe the changes of the kinetic properties when the composition changes from lean to reach mixtures.

4 Conclusions

A kinetic study of the catalytic combustion of stagnant *n*-butane/air mixtures on an isothermally heated platinum wire was undertaken to establish the effect of composition, total pressure and catalyst temperature. The reaction rates were measured for different mixtures containing 1.0, 1.5, 2.0, 2.5, 3.12, 4.0 or 4.5% fuel in air or in oxygen, at various total pressures between 10 and 100 kPa and temperatures between 680 and 1,130 K, significantly higher than the ignition temperatures. The obtained data were used to evaluate the overall and partial reaction orders on the basis of an empirical power law kinetic equation. The overall reaction order varies between 0.20 and 0.42 without an apparent trend with respect to fuel concentration or catalyst temperature. It indicates an important increase of the reaction rate with the total pressure at constant molar fractions of the components. Within the limits of experimental errors, the reaction order with respect to *n*-butane was close to unity, in agreement with other reported data. The reaction order with respect to oxygen was negative, around -0.16, indicating its inhibiting effect within this range of experimental conditions. The pressure dependence of the reaction rate was analyzed using the empirical power law and two mechanistic kinetic equations based on the Langmuir–Hinshelwood model, implying a non-dissociative or a dissociative mechanism of oxygen adsorption. The discrimination between these models on the basis of statistical analysis was not possible. However, both models are able to explain the quasi-hyperbolic reaction rate dependence on the total pressure. The overall activation energy obtained on the basis of Arrhenius equation is characteristic for a diffusion controlled process, while the pressure dependence of the reaction rate indicates a mixed control. The turnover frequencies of the order 10^3 s^{-1} prove the high efficiency of platinum for the catalytic combustion of *n*-butane.

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