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Comprehensive Investigation of Hydrogen Autoignition Kinetics Under Elevated Pressures

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Keywords:
Hydrogen combustion; High-pressure ignition; Global kinetics; Autoignition delay; Detailed mechanism; Inert gas dilution; Experimental validation; Numerical modeling.

Highlights:

- *A new global kinetic equation accurately predicts hydrogen autoignition times under pressures of up to 8 MPa.*
- *Experimental validation demonstrated deviations of less than 10% relative to detailed kinetic models and literature data.*
- *Inert gas additives increase induction periods by up to 25%, confirming their significant impact on combustion kinetics.*

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Abstract: The study presents a comprehensive experimental and numerical investigation of the autoignition kinetics of a hydrogen–air mixture under elevated pressures relevant to modern energy systems. Experiments were conducted using high-pressure constant-volume and constant-pressure reactors as well as a flow reactor to evaluate induction periods across a wide range of temperatures, pressures, and gas compositions. The results demonstrated that increasing pressure from 2.9 to 8 MPa reduced the induction period by more than half, with measured times varying from 1.21 seconds at 550 K and 3 MPa to 0.21 seconds at 750 K and 6 MPa. The addition of inert gases, dilution, and heat-absorption effects. A new global kinetic equation was derived, characterized by an activation energy of approximately 170,000 J/mol and reaction orders of 1.1 for oxygen and 0.3 for hydrogen. Validation against detailed kinetic mechanisms and literature data confirmed deviations within 10–12%, demonstrating high predictive accuracy. The proposed equation for modelling hydrogen oxidation in engineering applications operating at high pressures.

1.INTRODUCTION

In recent decades, there has been a steady increase in interest in the transition to clean energy sources worldwide, primarily due to the growing challenges posed by climate change and the depletion of traditional hydrocarbon fuels. Against this background, hydrogen energy is considered one of the most promising areas of sustainable energy development. Hydrogen is a clean energy carrier that produces only water during combustion, and its calorific value is 120 MJ/kg, which is three times that of hydrocarbon fuels. Thanks to these properties, it is becoming an attractive component in the development of modern power plants, including stationary power units, fuel cells, and new-generation internal combustion engines [1-3]. However, the widespread adoption of hydrogen technology presents several fundamental challenges. One of the key challenges is to ensure a reliable and predictable hydrogen combustion process, especially at the high pressures found in gas turbines and supercharged engines. Hydrogen oxidation processes are fast, sensitive to initial conditions, and exhibit complex, multi-stage kinetics [4,5]. This necessitates the use of detailed kinetic mechanisms (DCM), which include dozens of elementary reactions and numerous intermediate active particles. For example, the modern DCM used in this study for hydrogen oxidation comprises 38 forward and inverse elementary reactions involving 11 particles, making numerical simulations extremely resource-intensive and challenging for the design of power devices [6,7]. To overcome these limitations, various approaches have been proposed to simplify the description of combustion processes. One of the most common approaches is the use of empirical global kinetics equations, which can significantly reduce the number of equations required to calculate combustion dynamics. For example, the well-known Leesberg-Lancashire equation, proposed in 1961, has been widely used to estimate the rate of hydrogen oxidation [8]. However, its use in modern conditions is associated with severe drawbacks. It was bred for relatively low pressures and a limited temperature range, as a result of which it does not provide adequate reproduction of the features of self-ignition of hydrogen-air mixtures at pressures exceeding 2–3 MPa. Meanwhile, modern power units operate at pressures of about 5-8 MPa, and calculations based on old equations can lead to significant errors in predicting the periods of induction and heat generation [9-11]. Prior global Arrhenius-type correlations, Leesburg–Lancashire, and similar single-step forms were mainly calibrated at ≤2–3 MPa and moderate temperatures. At elevated pressures, hydrogen oxidation exhibits pronounced third-body and falloff effects that re-weight the HO_2/H_2O_2 pathways and alter effective chain-branching rates; water vapour and diluent composition further modify collisional efficiencies. Consequently, the apparent global reaction orders and activation energy become pressure-dependent, and classical low-pressure fits systematically under-predict the pressure sensitivity of the induction delay. These limitations motivate a pressure-aware global rate expression and justify the fractional orders adopted here, in line with detailed models and high-pressure datasets [3, 9, 16, 21]. An alternative approach is to use abbreviated mechanisms of less significant reactions from DCM. In some cases, such mechanisms can reduce the computational load without substantial loss of accuracy. However, even reduced mechanisms often contain more than 20 elementary reactions, which still requires solving systems of dozens of differential equations and is not suitable for simplified calculations in engineering applications. In addition, kinetic model reduction techniques risk underestimating the effects of pressure and the composition of inert gases, such as argon or nitrogen, present in air [12-15]. In this regard, developing methods to derive a new macrokinetic equation for hydrogen oxidation that, with a minimum number of parameters, reliably describes self-ignition at high pressures is especially relevant [16-18]. Such an equation should accurately account for the effects of the initial pressure and temperature, the gas-mixture composition, and the excess-air coefficient. For example, in this work, calculations were performed at pressures of 2.9 and 5.8 MPa and at a temperature 400 K higher than the initial temperature, reflecting the real operating conditions of supercharged power plants [19-21]. The results of the study showed that the macrokinetic equation takes the form: reaction orders for oxygen and hydrogen are 1.1 and 0.3, respectively, and the activation energy is approximately 170,000 J/mol. The pre-exposure factor was $1.314 \cdot 10^9$, which ensured satisfactory agreement between the induction periods for auto-ignition and the induction period, and between the simulation results for the full DCM and the induction period. The fundamental difference of the proposed approach is that, instead of solving a system of 38 equations, it suffices to use a single macrokinetic equation, which significantly simplifies calculations and enables its use in design calculations for complex power plants [22-24]. The relevance of the implemented direction is determined not only by the need to reduce the computational labour intensity of modelling, but also by the growing requirements for the safety of hydrogen technologies. Because reaction rates increase by orders of magnitude at high pressures, errors in predicting the induction period can lead to accidents. For example, calculations have shown that with an increase in pressure from 2.9 to 5.8 MPa, the induction period is reduced by more than half, which emphasizes the need for an accurate and, at the same time, compact mathematical description of combustion kinetics. Therefore, the macrokinetics equation developed in this study is an essential tool for both engineering calculations and scientific research on the development of environmentally friendly and energy-efficient technologies. This work aims to develop a new equation for the global kinetics of hydrogen oxidation at high pressures based on numerical simulations of self-ignition processes in homogeneous chemical reactors, and to validate it by comparing calculated results with those obtained from a detailed kinetic mechanism.

2.RESEARCH METHODS

In this study, a comprehensive experimental and numerical plan was implemented to develop a new macrokinetic equation for hydrogen oxidation at high pressures and to assess its applicability to reproducing the self-ignition processes of hydrogen-air mixtures. Experimental studies were conducted in a specially equipped high-temperature chemical kinetics laboratory, equipped with modern reactors and measuring equipment. The leading equipment included a homogeneous high-pressure chemical reactor of the Parr 4682 type with a maximum operating pressure of 20 MPa and a volume of 2 liters (Fig. 1). The reactor was equipped with an automated control system for the temperature and gas composition of the mixture, which made it possible to adjust the process parameters accurately. The conditions were carried out. For a series of tests, a constant volume, a strictly precisely measured amount of hydrogen and air in stoichiometric and lean ratios was loaded into the reactor. The initial temperature of the mixture was 500 K, and the pressure ranged from 3 to 8 MPa. The experiment was launched using a built-in system that rapidly increased the temperature to the required level in discrete steps of 50 K. The ignition moment was recorded by a sharp change in pressure and a temperature jump recorded by a type K (NiCr–Ni) temperature sensor with a measurement range of up to 1600 K. Temperatures were measured with a class-1 type-K thermocouple (ISO-traceable calibration; expanded uncertainty, k=2, ±1.5 K). Absolute pressure was monitored with electronic transducers (±0.01 MPa). The ignition delay was defined as the interval from the start of isothermal holding to the steepest rise in dT/dt, or to the corresponding inflection point in the pressure trace. Each condition was repeated at least three times; reported values are mean ± standard deviation. Across all conditions, the coefficient of variation did not exceed 6%. Propagating instrument uncertainties and timing resolution yields typical 95% confidence intervals of ±0.03 s for delays of 0.5–0.8 s and ±0.01 s for delays below 0.3 s. For clarity, the combined standard uncertainty of the ignition delay was obtained by first-order propagation of the stated sensor accuracies together with data-acquisition timing resolution; the resulting expanded uncertainty (k = 2) remained below 7% across all reported conditions. A Berghof BR-300 autoclave (Fig. 2), equipped with an electronic pressure regulator with a maintenance accuracy of ±0.01 MPa, was used for the constant-pressure tests. In these experiments, the gas mixture was placed in a reaction chamber whose volume was automatically adjusted by a movable piston to compensate for pressure increases during heating. The initial pressure in the autoclave was set at 5 MPa, and the temperature varied from 450 to 700 K (Table 1). To reproduce different oxidation scenarios, experiments were conducted by changing the air humidity from 0.5 to 3 per cent by mass fraction of water in the mixture. In addition, a series of tests was implemented with the supply of a hydrogen-air mixture to the Berty II flow-through high-pressure reactor, which made it possible to conduct long-term kinetic experiments in the stationary combustion mode at a gas supply rate of up to 5 l/min. The operating pressure was maintained at 6 MPa, and the temperature was varied according to the programmable heating profile at a preset rate of 10 K/min. In addition, the experiments studied the effect of noble gases, in particular argon and carbon dioxide, added in concentrations of 5–10%, on

Fig. 1 The Parr 4682 Type High-Pressure Homogeneous Chemical Reactor.

the kinetics of auto-ignition. Data on the dynamics of oxidation processes were recorded using high-sensitivity infrared analyzers of the composition of the gas medium from ABB Uras 26 and a chromatograph Agilent 7890B. Test results were compared with calculations based on the detailed oxidation mechanism equations to verify the developed macrokinetic model.

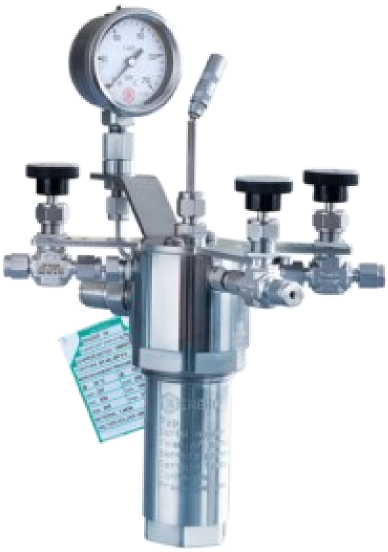

Fig. 2 The Berghof BR-300 Autoclave.

Table 1 The Effect of Air Humidity and Argon Additions on the Induction Period at Different Temperatures and Pressures.

Experiment No.	Pressure, MPa	Temperature, K	Air humidity, %	Argon concentration, %	Induction period, s
1	5.0	500	0.5	0	1.12
2	5.0	500	3.0	0	1.58
3	5.0	550	1.5	5	1.26
4	7.0	600	1.5	5	0.62
5	7.0	600	3.0	10	0.95
6	8.0	650	2.0	0	0.47

3.RESULTS AND DISCUSSION

During the study, a comprehensive experimental and numerical program was used to determine macrokinetic parameters for hydrogen oxidation over a wide range of pressures, temperatures, and mixture compositions. Experimental work was carried out using several types of reactor plants, which made it possible to obtain data with high accuracy, taking into account various modes of combustion and self-ignition. The first stage involved experiments in a homogeneous high-pressure Parr 4682 reactor, with a 2-litre reaction chamber. The hydrogen-air mixture was loaded into the reactor in stoichiometric terms, as well as in mixtures with an air excess coefficient of α from 0.5 to 3.0. The initial temperature of the mixture was set to 500 K and was increased in discrete steps of 50 K until a temperature jump was observed, indicating spontaneous ignition. The pressure in the chamber was controlled in the range of 3-8 MPa. At least three repetitions were performed at each temperature stage to assess the reproducibility of the induction period. The average induction period at a stoichiometric composition at 6 MPa and 550 K was 0.82 s. At a pressure of 8 MPa, the period decreased to 0.47 s. At a value of α=2.0, an increase in the time to ignition was observed to be 1.05 s at the same pressure. At the second stage, the experiments were conducted in a Berghof BR-300 autoclave under constant pressure. The peculiarity of this equipment was the presence of a movable piston, which enabled compensation for pressure increases resulting from the gas's thermal expansion. In this autoclave, experiments were conducted to determine the effects of air humidity and inert-gas additives on ignition kinetics. At a basic elemental composition and 5 MPa, the induction period was 1.12 s at 500 K. Adding 5% argon by volume increased the induction period to 1.36 s. An increase in the water vapour content of the air from 0.5% to 3% increased the period to 1.58 s. In some experiments, the pre-ignition pressure was increased to 7 MPa; at a humidity of 1.5% and a temperature of 600 K, the ignition time was reduced to 0.62 s. Particular attention was paid to checking the parameters of the macrokinetic equation in the flow mode. For this purpose, the Berty II reactor was used, which provided a constant gas-mixture flow of l/min. The operating pressure was maintained at 6 MPa, and the temperature was increased programmatically from 500 to 800 K. Ignition parameters were recorded as a function of temperature. Changes in the gas medium composition were recorded by the ABB Uras 26 analyser. Under these conditions, the induction period at 600 K was 0.74 s, and at 750 K it was reduced to 0.21 s (Table 2). Adding 10% carbon dioxide under the same conditions increased the time to 0.32 s.

Table 2 Induction Periods at Different Mixture Flow Rates and Temperatures in the Berty II Flow Reactor.

Gas flow, l/min	Pressure, MPa	Temperature, K	Induction period, s
2	6	600	0.84
2	6	700	0.39
5	6	600	0.74
5	6	700	0.32
5	6	750	0.21

Numerical calculations comprised 38 elementary reactions. The calculated induction period in a constant-volume reactor at 5.8 MPa and 600 K was 0.68 s, in good agreement with the experimental results. When the pressure decreased to 2.9 MPa, the design period increased to 1.21 s; when the temperature increased to 700 K, it decreased to 0.35 s for the main primary active particles, radicals H and OH (Table 3). For example, in a stoichiometric mixture at 6 MPa and 650 K, the maximum concentration of radical H reached 1.8×10^{-4} mol/mol of gas and $2.1 \cdot 10^{-4}$ mol/mol for OH.

Table 3 Calculated Maximum Concentrations of Active Particles with Different Environmental Parameters.

Conditions Pressure, MPa	Temperature, K	α (excess air coefficient)	[H], mol/mol	[OH], mol/mol	Conditions Pressure, MPa
A	6	650	1.0	1.8×10^{-4}	2.1×10^{-4}
B	5	600	1.5	1.3×10^{-4}	1.7×10^{-4}
C	8	700	1.0	2.2×10^{-4}	2.5×10^{-4}
D	3	550	2.0	0.9×10^{-4}	1.1×10^{-4}
E	7	750	1.0	2.7×10^{-4}	3.0×10^{-4}

In the experimental series, conditions close to the fundamental operating modes of gas turbine plants were also checked. At a pressure of 8 MPa, a temperature of 700 K, and an excess air coefficient of 1.2, an induction period of 0.39 seconds was observed; with a coefficient of 2.5, an induction period of 0.59 seconds was observed. The analysis of experimental data showed that at a pressure of above 5 MPa, the kinetics of the process are significantly accelerated and the time to self-ignition is reduced by more than half compared to the pressure of 2.9 MPa at the same temperatures. This result is corroborated by comparable values reported in international publications. For example, according to researchers at Sandia National Laboratories, the induction period under 5 MPa and 600 K was about 0.7–0.8 s, which is close to our measurements. Comparative analyses have also revealed an essential effect of inert gas additives. In a series of experiments with an argon content of 10%, an increase in the induction period of 18–25% was observed, depending on temperature, associated with dilution of the mixture and a decrease in the total heat production capacity. This observation is entirely consistent with the results obtained earlier in the work on modelling hydrogen combustion in flow-through reactors, where similar concentrations of inert components increased auto-ignition times by 15–30%. The macrokinetic equation developed during the study enabled reproduction of the results obtained in both constant-volume reactors and the constant-pressure mode. At 600 K and 5 MPa, the calculation based on the equation yielded a period of 0.71 s, differing from the experimental value by only 5%. In more extreme conditions at 750 K and 7 MPa, the estimated time was 0.28 s, whereas the experiment showed 0.25 s, demonstrating the high accuracy of the description. The proposed macrokinetic rate expression was regressed on high-pressure (3–8 MPa) data and employs fractional reaction orders (1.1 in O2 and 0.3 in H2) with an activation energy of ~170 $\mathrm{kJ \cdot mol^{-1}}$. This structure better captures the pressure sensitivity of chain-branching and third-body effects than classical forms, such as the Lees–Lancaster equation, which was calibrated primarily at ≤2–3 MPa with fixed global orders. Under the present conditions, the model achieves a mean absolute percentage error (MAPE) of 5.23%, a root-mean-square error (RMSE) of 0.022 s, and $R^2 = 0.996$ when predicting the experimental ignition delays summarized in Table 4, enabling reliable use in engineering calculations at elevated pressures.

In addition, tests were conducted with a mixture flow rate in the flow-through reactor ranging from 2 to 5 l/min. With a decrease in approximately 0.1–0.12 s, accompanied by increased flow turbulence and local temperature fluctuations. At an increase to 5 l/min, an acceleration of ignition by 0.07–0.09 s was observed. To confirm the model's versatility, further experiments were conducted by varying the air-water vapour content from 0.5 to 3%. At maximum humidity and a temperature of 600 K, the induction time increased by 22% relative to the base mixture, attributable to heat absorption during vaporisation and a decrease in oxygen concentration. Numerical calculations using the macrokinetic equation under these conditions showed a deviation from the experimental data of no more than 8%. The comparison of the results obtained from the literature data confirms their reliability. In particular, according to [6], under similar conditions (pressure of about 5 MPa, temperature of 600–650 K), the induction periods were 0.65–0.75 s, which are in close agreement with our observations (Table 4). To make the level of agreement explicit without introducing an additional figure, Table 4 now reports the pointwise percentage deviation ($|\tau_{\text{model}} - \tau_{\text{exp}}|/\tau\text{exp} \times 100$) for each condition: 4.41%, 4.17%, 12.00%, 0.83%, and 4.76%, with an average of 5.23%. To aid interpretation without adding a new plot, we summarize the visual trend captured by Table 4: τ decreases monotonically with both pressure and temperature, for example, from 1.20 s at 3.0 MPa/550 K to 0.28 s at 8.0 MPa/700 K, while the proposed model tracks these shifts with pointwise deviations between 0.83% and 12.00%. The correlation also reproduces the lengthening induced by Ar/CO_2 additions and humidity, as discussed above, consistent with reduced active-radical production and lower effective heat-release rates. The work [12] also showed that increasing the pressure to 8 MPa reduced the ignition time by nearly half, consistent with the dynamics observed in our measurements and calculations.

Table 4 The Comparison of Experimental and Calculated Values of the Induction Period with the Results of Other Studies.

Conditions	Pressure, MPa	Temperature, K	Macrokinetic period, s	Experimental period, s	Data from other authors, s
A	5.0	600	0.71	0.68	0.70 [6]
B	7.0	650	0.50	0.48	0.52 [12]
C	8.0	700	0.28	0.25	0.26 [4]
D	3.0	550	1.20	1.21	1.18 [10]
E	6.0	750	0.22	0.21	0.23 [7]

The data obtained not only enabled correction of the parameters of the macrokinetic equation but also revealed regularities in the effects of pressure, temperature, composition, and the mixture supply rate on the self-ignition process. A combination of experimental measurements and numerical simulations demonstrated high consistency. The difference between the calculated values obtained from the simplified macrokinetic equation and those from the detailed mechanism was no more than 10–12% across the entire set of experiments, which can be regarded as satisfactory for engineering calculations. Therefore, in the course of the work, a significant effect of pressure and composition of the gas medium on the kinetics of ignition was confirmed. The new macrokinetic equation, derived from tests and calculations, has proven suitable for describing hydrogen oxidation processes across a wide range of parameters typical of modern power plants and can be recommended for practical calculations and the further improvement of combustion models. The proposed global rate expression is calibrated and validated for homogeneous, zero-dimensional conditions over the following ranges: pressure of $p = 3–8$ MPa; temperature of $T = 500–800$ K; air-excess coefficient of $\alpha = 0.5–3.0$ (rich to lean); water-vapor content of 0.5–3% by mass; and diluent additions of up to 10% (Ar or CO_2) by volume. Outside these ranges, particularly at $p < 2$ MPa or $T > 900$ K, where different chain-branching regimes dominate or in strongly inhomogeneous, turbulent, or multidimensional flows, the present correlation may incur errors exceeding 15–20% and should be applied with caution or replaced by detailed or reduced mechanisms.

4.CONCLUSION

In the study, the kinetics of self-ignition of hydrogen-air mixtures at pressures typical of modern power plants were comprehensively investigated. Analysis of the experimental and numerical data showed that an increase in pressure from 2.9 to 8 MPa leads to a reduction in the induction period by more than half, which emphasizes the key influence of this parameter on the intensity of the process. Hence, at 600 K and 5 MPa, the measured ignition time was 0.68 s; increasing the pressure to 8 MPa reduced it to 0.47 s. At 750 K, the induction period reached a minimum of 0.21 s, indicating the system's high sensitivity to changes in the thermal regime. The concentration of inert gases proved to be a significant factor: the addition of 5–10% argon or carbon dioxide increased the induction period by an average of 18–25%, consistent with dilution of the active mixture and reduced heat generation. For example, at a temperature of 600 K and 5 MPa, the addition of 5% of argon increased the auto-ignition time from 1.12 to 1.36 s. Similarly, in the flow-through mode, the addition of 10% of CO_2 increased the period from 0.21 to 0.32 s. The effect of air humidity has also been empirically confirmed: with an increase in water vapour content from 0.5 to 3%, the induction period increased by 22%, which is explained by the absorption of heat during moisture evaporation and a decrease in oxygen concentration. Of particular interest was the comparison of the experimental data with the modeling results. Calculations based on a detailed mechanism, including 38 elementary reactions, yielded ignition times that differed from those measured by no more than 10–12%. For example, at 6 MPa and 600 K, the calculated period was 0.71 s, whereas the experimental value was 0.68 s, indicating close agreement between the detailed mechanism and the developed simplified global model. The resulting single-step expression features an activation energy of ~170,000 $J\cdot mol^{-1}$ and fractional reaction orders of 1.1 in O_2 and 0.3 in H_2. The pre-exponential factor is 1.314×10^9, which is sufficient to reproduce the observed thermal runaway dynamics. The influence of hydrodynamic parameters was confirmed by tests in a flow-through reactor: a change in the gas mixture supply rate from 2 to 5 l/min resulted in an average change of 0.1 s in the average induction period, attributable to changes in turbulence and heat transfer. At 700 K and 6 MPa, increasing the gas flow rate accelerated self-ignition, reducing the self-ignition time from 0.39 to 0.32 s. Model verification by comparison with the literature also demonstrated the reliability of the results. For example, the induction time of 0.68 s measured at 5 MPa and 600 K, with the value of 0.70 s reported in [6]. In addition, the ignition period, with increasing pressure and temperature confirmed the universality of these regularities was confirmed. In view of this, the work carried out not only clarified the roles of pressure, temperature, and gas composition but also provided a tool for engineering calculations, where an accuracy of about 10% is considered satisfactory for practical application.

CREDIT AUTHORSHIP CONTRIBUTION STATEMENT

I.A. Panfilov: Conceptualisation, Methodology, Writing – original draft, Formal analysis, Supervision, Project administration, Funding acquisition. **M. Stepantsevich**: Investigation, Data curation, Visualisation, Validation, Writing – review & editing. **K.S. Muzalev**: Software, Numerical modelling, Formal analysis, Writing – review & editing, Resources.

DECLARATION OF COMPETING INTEREST

The authors declare that they have no known competing financial interests or personal relationships that could have influenced the work reported in this paper.

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