

HIGH-TEMPERATURE IGNITION OF HYDROGEN AND AIR AT HIGH PRESSURES DOWNSTREAM OF THE REFLECTED SHOCK WAVE

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The delay and features of the ignition of a stoichiometric hydrogen–air mixture at temperatures of 830–1450 K and pressures of 2–21 atm downstream of the reflected shock wave have been investigated. The experimental data obtained were compared with the results of calculations done in accordance with the known kinetic schemes of hydrogen combustion. The influence of the interaction of the reflected shock wave with the dynamic boundary layer on the ignition and the time of induction in a shock tube has been examined.

Introduction. The critical conditions and mechanisms of hydrogen ignition, which are characterized by a number of significant features, are fairly well understood. These features are explained in the theory of chain reactions by the competition of the reactions of branching and interruption of chains [1–4]. The most important of these features are the existence of three thresholds of chain hydrogen ignition at certain initial parameters of the reaction mixture [2] and the complex functional dependence of the ignition delay or the induction time on the pressure and temperature [5].

The kinetics of high-temperature hydrogen oxidation ($T > 800$ K) at low pressures (lower than 0.2 MPa) has been thoroughly studied and verified with the use of a large amount of experimental data on this process in mixtures highly diluted with argon downstream of the incident and reflected shock waves. At these parameters the ignition delay, as a rule, is inversely proportional to the partial pressure of oxygen in the original mixture and decreases with increase in the initial pressure. In this case, rapid chain reactions with the participation of H, OH, and O radicals [1–4] are dominating among the chemical reactions occurring in the mixture.

An increase in the pressure of the mixture (>0.2 MPa) increases the role of the chain branching reactions with the participation of H_2O molecules formed in the trimolecular reaction $H + O_2 + M = HO_2 + M$ and favors the removal of active hydrogen atoms from the rapid cycle reactions determining the ignition at low pressures. The branched cycle, in which H_2O molecules are formed, consists of a number of slow reactions decreasing the characteristic time of oxidation of the mixture and the ignition delay. The numerical values of the induction time at different initial parameters of the mixture (P, T), the knowledge of which is extremely important for many practical applications, are usually determined through the numerical integration of a system of kinetic equations [6, 7] or through the use of empirical temperature approximations obtained on the basis of measurements at low pressures. The inaccuracy of the rate constants of chemical reactions or of other kinetic parameters, which are not clearly understood, badly influences the results of calculations and, every so often, decreases the reliability of the prediction of the delay of ignition of hydrogen as a result of its oxidation on the basis of the extrapolation of known reaction mechanisms and their approximation for the case of high pressures. Moreover, the existing kinetic schemes of hydrogen oxidation are not supported by detailed experimental data on this process at high pressures. Few works [8–10] were devoted to investigation of high-temperature hydrogen ignition at pressures of 2–9 atm downstream of the incident and reflected shock waves, and the delay of ignition in a mixture of hydrogen with air was measured only in one of these works [10].

Thus, the range of pressures 2–20 atm and temperatures 900–1400 K, characterized by a change in the mechanism of hydrogen oxidation and a nonmonotonic dependence of the induction time on the temperature and pressure, was practically not investigated. At the same time, the prospects of wide use of hydrogen in power engineering, machine building, and in a number of other modern industries call for a more detailed knowledge of the features of the combustion of hydrogen with air at high pressures. Therefore, the aim of the present work is experimental determina-

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TABLE 1. Ranges of the Initial Parameters of the Ignition of a Stoichiometric Hydrogen–Air Mixture Downstream of the Reflected Shock Wave

Mixture	P , atm	T , K
1	2.1–3.86	890–1433
2	5.1–7.72	989–1385
3	6.97–9.7	980–1360
4	8.25–11.2	975–1365
5	10.1–4.9	949–1359
6	14.3–20.4	835–1203

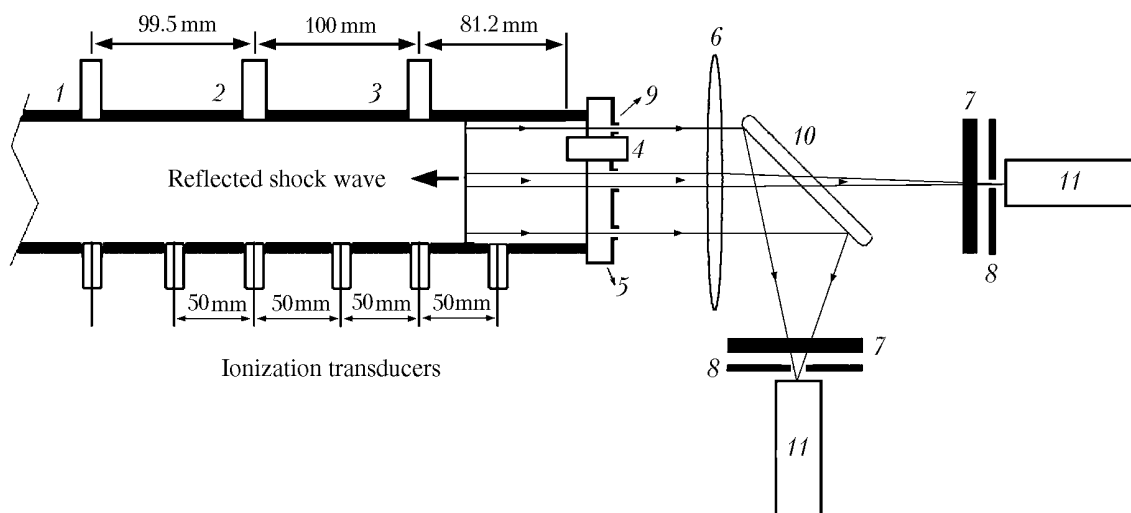


Fig. 1. Diagram of the measurement unit of a shock tube: 1–4) high-frequency pressure transducers; 5) transparent reflection face of the shock tube; 6) lens ($f = 40$ cm); 7) interference filters; 8) diaphragms; 9) ring stop; 10) beam-splitting plate; 11) photomultipliers.

tion of the kinetics of ignition of hydrogen–air mixtures at pressures of up to 20 atm and temperatures of 900–1400 K and verification of the validity of predictions of detailed kinetic mechanisms of hydrogen oxidation in the case of high pressures.

Experimental Setup. For measurements of the delay of ignition of a hydrogen–air mixture downstream of the reflected shock wave we used a shock tube of stainless steel with a diameter of 50 mm and a length of 8.5 m. Experiments were conducted with a stoichiometric hydrogen–air mixture at pressures of $P = 3$ –21 atm downstream of the reflected shock wave. The compositions of the mixtures studied and the thermodynamic conditions under which they were investigated are given in Table 1. The gas mixtures were prepared from chemically pure hydrogen (99.99%) and air compressed at a high pressure by the manometric method and were allowed to settle for several days before their use. Prior to an experiment, the tube was treated two times under vacuum to a residual pressure of $\sim 10^{-2}$ mm Hg. To exclude the influence of parasitic mixtures on the composition of the gas in the low-pressure chamber of the shock tube, we cleaned the inner channel of the tube by the mixture studied prior to the second vacuum treatment. The initial pressure in the shock tube was controlled with the use of a standard pressure gauge with an accuracy of ± 0.3 mm Hg.

The dynamics of change in the pressure in different cross sections was recorded with the use of PCB Piezotronics piezoelectric transducers calibrated in advance with a spatial resolution of no less than 1.5 mm (Fig. 1). The transducers measured the pressure at a signal build-up time of more than 1 μ sec. The gas parameters were calculated with the use of a shock adiabat by the measured velocity of the incident shock wave. The velocity of the incident shock wave was determined by the time of its arrival at the pressure transducers positioned in different cross sections of the measurement unit of the shock tube. The primary recording of the signals was performed by six-channel, ten-

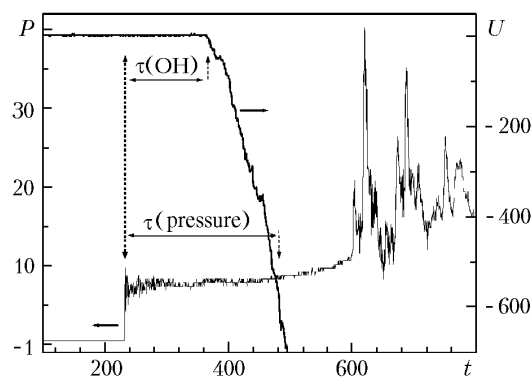


Fig. 2. Oscillograms of the change in the glow of the OH radical and in the pressure at the face of the shock tube as a result of the ignition of hydrogen downstream of the reflected shock wave.

digit oscillographs. The results of the measurements were analyzed and the thermodynamic parameters of the medium were calculated with the use of an automated system for collection and processing of experimental data.

The self-ignition of the mixture was observed through a transparent optical window attached to the face of the shock tube (Fig. 1). A piezoelectric transducer was installed in the frame of the window near the axis of the channel of the shock tube. The indications of the transducer were used for measuring the time of reflection of the wave and the evolution of the pressure downstream of the reflected shock wave near the face of the shock tube. For the purpose of detection of the onset of ignition, the radiation from a volume of diameter 5 mm located at the shock-tube axis was directed through a field stop onto the cathode of a photomultiplier. An aperture stop of diameter 0.5 mm positioned at the focus of a collecting lens (Fig. 1) provided spatial filtration of the received radiation and transmitted only those light beams that propagated along the shock-tube axis. To estimate the influence of the reflected shock-wave bifurcation on the hydrogen ignition, we directed the radiation from a ring layer of outside diameter 44 mm and inside diameter 38 mm, located in the near-wall region, onto another photomultiplier through a ring stop installed on the outer side of the reflecting wall (Fig. 1). A beam-splitting plate divided the output radiation and, in doing so, formed two optical paths that were used for simultaneous recording of the spectrum in different parts of the shock tube. Monochromatic interference filters transmitted only the needed spectral radiation. The ignition delay was measured by the radiation of the OH radical ($\lambda = 306.2$ nm, $\Delta\lambda = 2.4$ nm) and additionally controlled by the beginning of increase in the gas pressure at the shock-tube face and the instant the flame front arrived at the ionization transducers installed along the measuring unit.

The ignition delay (induction time) was determined as the time interval between the instant the shock wave begins to reflect and the instant a glow of needed intensity appears in the volume of the reacting mixture. This measurement scheme made it possible to detect the onset of self-ignition at any site of the observation region and determine the absolute induction time from the instant the incident shock wave begins to reflect.

The intensity of the proper glow, by which the ignition delay was estimated in the channels for emission measurements, was determined in the following way:

1. The regime of intense ignition of hydrogen (hard regime) at a temperature of higher than 1200 K downstream of the reflected shock wave was initiated.

2. Under these conditions, a volume explosion of the combustion mixture with a small induction time was realized, with the result that a stationary detonation wave was formed even at a distance of 81.2 mm from the reflecting face. The characteristic oscillograms of the glow and pressure in the case of hydrogen ignition downstream of the reflected shock wave are shown in Fig. 2.

3. From a glow oscillogram we determined the change in the intensity of the proper glow in the spectral range ΔU for the time Δt (15–30 μ sec as a rule) of propagation of a stationary detonation.

4. Then the intensity of the glow of the reaction zone of thickness 1 mm was calculated by the simplest relation $U = \Delta U / (V_{32} S \Delta t)$.

5. The quantity U was divided by the density of the gas downstream of the reflected wave: U/ρ^* .

6. A glow intensity equal to $(U/\rho^*)\rho$ was used for determining the induction time of the working mixture.

The ignition delay determined in this way can be assigned to the instant of maximum energy release at the ignition site of diameter 5 mm formed at the axis of the shock tube in the volume of the shock-compressed gas downstream of the reflected shock wave. In the case where the induction time was measured in the ring layer located in the near-wall region, the intensity of the proper glow was recalculated by the ratio between the areas of the field stops (see Fig. 1).

To determine the initial stage of self-ignition, we also estimated the induction time of the mixture by the instant the signal of the OH radical glow begins to increase. This criterion was used for analysis of the kinetics of chemical reactions in the preinduction period.

In the course of experiments, the temperature of the mixture varied from 850 to 1400 K at a constant density of the gas downstream of the shock wave. This made it possible to maintain the concentrations of the combustible, oxygen, and nitrogen at a constant level within the measurement error at all the temperatures studied.

Measurement Errors. The main experimental errors are explained by the following facts. First, the velocity of the incident shock wave was measured with an error of 0.5%, which led to a 0.5–1.5% error in the determination of the thermodynamic parameters of the mixture (T , P) downstream of the reflected shock wave. Second, the criteria for determining the ignition delay were selected in a somewhat arbitrary way. According to our estimations, this could cause a 10–20% error in the measurement of the induction time for all the hydrogen–air mixture temperatures studied.

Numerical Simulation of Ignition. To verify the exactness of the detailed mechanisms of the chemical reactions occurring in the ignition of a hydrogen–air mixture, we have performed numerical simulation of the experiments on this process in shock tubes. In experiments on the kinetics of combustion, various nonideal effects of the gas flow are usually minimized to simplify the analysis of the data obtained [11]. There are also other factors that make the interpretation of the results of experiments on the ignition in shock tubes difficult; for example, the pressure, size, and temperature of both the reacting gas volume and the surrounding gas change with time. Therefore, the adjacent elements of the shock-heated and compressed gas have different reaction orders, and in the mixture downstream of the shock front there are always spatial gradients of the density, temperature, and composition, which can significantly influence the ignition processes.

It was assumed that the nonideal effects have no influence on the experimental data and so they can be ignored in calculations. The process of high-temperature oxidation of hydrogen downstream of the shock-wave front was considered with the use of the mathematical model of adiabatic ignition of an ideal gas at a constant pressure, based on a widely used program packet [12]. Modern kinetic schemes [13–15] of processes including as much as 28 chemical reactions with the participation of hydrogen and its derivatives were used in the calculations. The calculated and experimental data on the ignition delay were compared by the maximum production of the OH radical in the reaction of hydrogen oxidation under conditions corresponding most closely to the experimental ones.

Results. The results of the measurements of the induction time for several values of the density of the hydrogen–air mixture downstream of the reflected shock wave are presented in Fig. 3. The induction time was measured by the glow of the region located at the axis of the shock tube and the glow of the region located in its peripheral part at a wavelength of 306.2 nm ($\Delta\lambda = 2.2$ nm; radical OH) as well as by the instant the combustion wave front arrives at the ionization transducers and the pressure at the reflecting wall. At high temperatures of $T > 1100$ K, the hard regime of hydrogen ignition was realized in the experiments [16, 17]. This regime is characterized by volume ignition of the mixture with a short induction time and practically instantaneous formation of a strong shock wave everywhere over the cross section of the shock tube channel. This wave, a short distance away, transforms to a detonation wave. It may be suggested that the detonation wave arises near the reflecting face since the time of arrival of the detonation front at the ionization transducer positioned at a distance of 31.2 mm from the reflecting wall was practically independent of the temperature (Fig. 3a and b). In the hard regime of ignition, the induction times determined by the glow of the OH radical in the region located at the axis of the shock tube and by this glow in the region located in its near-wall part were equal within the measurement error, which indicates that the gas ignition is homogeneous over the cross section of the shock-tube channel.

The results of the numerical simulation performed in accordance with the kinetic mechanism of A. A. Konnov (version 4.0) [13] and the GRIMECH 3.0 kinetic mechanism [15] correlate fairly well with the experimental data at temperatures of $T > 1100$ K. In this case, the best agreement is observed at low pressures (2.1–3.86 atm) of the hydro-

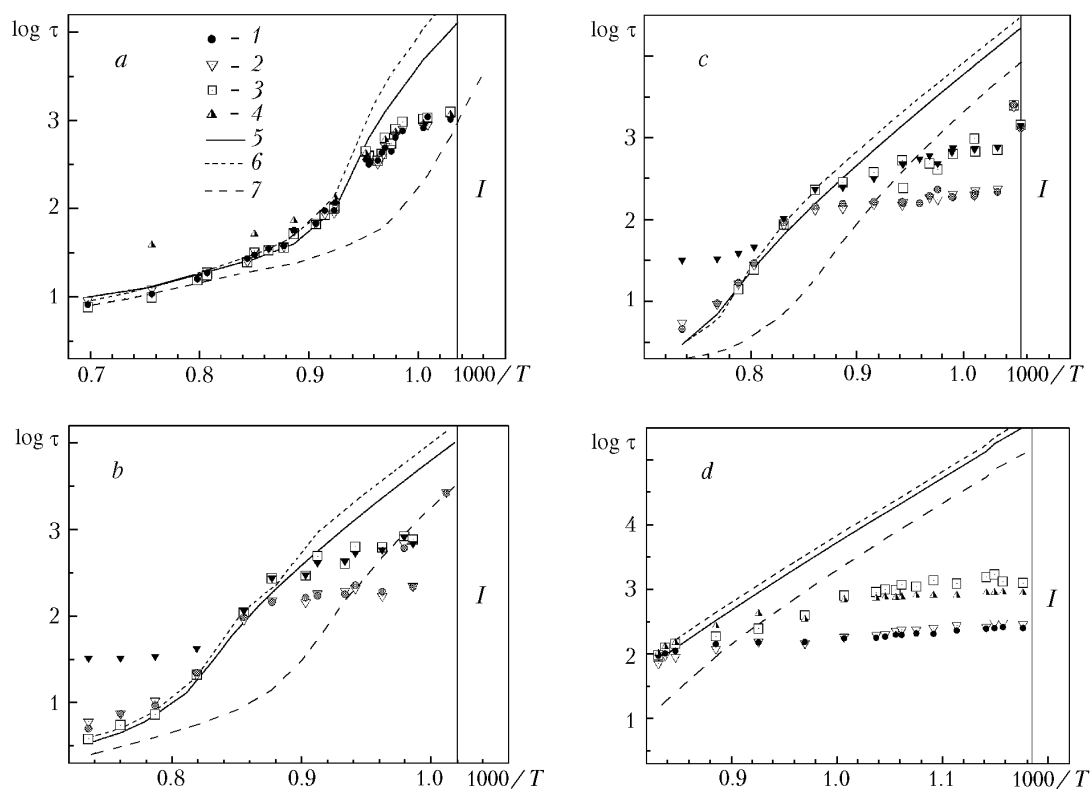


Fig. 3. Dependence of the induction time on the inverse temperature of a stoichiometric hydrogen–air mixture for different densities of the gas downstream of the reflected shock wave: a) mixture 1, $\rho = 0.67 \pm 0.02 \text{ kg/m}^3$; b) mixture 3, $\rho = 1.8 \pm 0.015 \text{ kg/m}^3$; c) mixture 5, $\rho = 2.8 \pm 0.03 \text{ kg/m}^3$; d) mixture 6, $\rho = 4.32 \pm 0.07 \text{ kg/m}^3$; 1, 2) measurements by the glow of the OH radical in the region located at the axis and in the near-wall region of the shock tube; 3) measurements by the pressure at the reflecting wall; 4) measurements by the instant the ignition front arrives at the ionization transducers; 5) calculation in accordance with the mechanism of [13]; 6) calculation in accordance with the mechanism of [14]; 7) calculation in accordance with the mechanism of [15]. *I*) Temperature range where hydrogen does not ignite downstream of the reflected shock wave.

gen–air mixture downstream of the shock wave (Fig. 3a). At temperatures of $T > 1250 \text{ K}$ and pressures of 7–15 atm, the calculations done in accordance with the mechanism of A. A. Konnov (version 4.0) and the GRIMECH 3.0 mechanism give induction times 1.2–1.5 times shorter than the measured ones. In this case, the data obtained by simulation of the ignition by the GRIMECH 3.0 [15] kinetic scheme are in better agreement with the experimental data (Fig. 3a–c). Thus, the description of the hydrogen oxidation kinetics at temperatures of $T > 1250 \text{ K}$ and pressures of $P > 7 \text{ atm}$ with the use of the above-indicated reaction mechanisms is not sufficiently adequate.

Significant systematic differences between the measurement and calculation data were observed at temperatures of $T < 1100 \text{ K}$, and these differences increased with increase in the pressure (Fig. 3c) and reached a maximum value of the order of 10^2 at a density of the mixture downstream of the reflected shock wave of $\rho = 4.32 \pm 0.07 \text{ kg/m}^3$ (Fig. 3d). At the same time, while the pressure and density of the gas increased, the induction time remained practically independent of the temperature of the mixture downstream of the reflected shock wave. As the temperature decreased in the experiments, the hard regime of hydrogen ignition changed to a mild one [16, 17] with a smooth increase in the pressure in the process of formation of local sites of self-ignition in the volume of the compressed, heated gas downstream of the reflected shock wave. In this case, a further increase in the temperature and pressure

led, as a rule, to an increase in the rate of the reaction and a thermal explosion in the surrounding fragments of the fresh mixture. Because of the change in the mechanism of hydrogen oxidation and, as a consequence, the decrease in the sensitivity of the mixture to the temperature changes, the evolution of the flow with increase in the pressure in the mild regimes was not accompanied by a marked acceleration of combustion and led to a small increase in the pressure of the residual gas downstream of the reflected shock wave. Under these conditions, the recording of the pressure at the face of the shock tube became insensitive to the instant of onset of ignition and gave substantially overestimated values of the induction time as compared to those obtained in the channels for emission observations (Fig. 3b–d).

The emission measurements have shown that both the regime of ignition and the temperature dependence of the ignition delay change at temperatures of $T < 1200\text{K}$ in the case where the glow of the OH radical in the peripheral region regularly appears earlier than this glow in the region located at the tube axis. Thus, when the regime changes, the hydrogen ignition becomes inhomogeneous over the cross section and appears earlier in the region located near the shock-tube wall. This tendency becomes most marked when the density of the hydrogen–air mixture increases (Fig. 3c and d) and is explained by the bifurcation of the reflected shock wave as a result of its interaction with the dynamic boundary layer [18, 19]. In the case of existence of a turbulent boundary layer on the wall of the shock tube, as in our experiments, where the Reynolds number Re determined by the diameter of the channel and the minimum velocity of the flow downstream of the incident shock wave was larger than $8 \cdot 10^5$, this leads to a splitting of the shock-wave front and a self-similar increase in the bifurcation structure consisting of a λ -shaped shock wave and a separation flow zone [20].

An important characteristic determining the ignition under these conditions is the local increase in the temperature of the mixture, as compared to the mean temperature downstream of the reflected shock wave, which is locally heated as a result of the interaction of the reflected shock wave with the turbulent boundary layer. It has been experimentally established in [20] that, as a result of the reflected shock wave bifurcation, the pressure reaches a peak value that is 10–15% higher than the pressure downstream of the reflected wave. This local excess can be explained by the fact that the stagnation pressure of the gas jet passed through two oblique shocks of the bifurcation zone is higher than the pressure at the flow core downstream of the reflected shock wave. However, the small pressure peak corresponding to a local increase in the mixture temperature by approximately 50–70 K does not explain the practically constant ignition delay at high densities of the hydrogen–air mixture in the wide temperature range 830–1200 K (Fig. 3c and d). One possible reason for this could be the interaction of the reflected wave with the region of transition to the turbulent boundary layer downstream of the incident shock wave. The existence of this transition region is explained by the fact that in the flow there rapidly arise nonconvergent responses having a definite wavelength, with the result that a vortex street is formed and the laminar boundary layer loses its stability. At the site of transition, the velocity and local thermodynamic parameters are very nonuniform. The diffraction of the reflected shock wave on these nonuniformities can lead to focusing of the shock wave and, as a consequence, to a large local increase in the temperature and density of the mixture as compared to their mean values in the flow. The hot sites formed are sources of firing of the mixture near the wall of the shock tube. In the range of pressures and temperatures of the gas downstream of the reflected wave, where the induction time near the face of the shock tube is smaller than that at the site of interaction of the reflected wave with a stream instability in the region of the transition, the ignition of the mixture in the bifurcation zone begins to significantly influence the measurement results and the further evolution of the flow.

It should be noted that, in the case where the laminar boundary layer loses its stability, the degree of nonuniformity of the flow increases with increase in the density of the gas downstream of the incident wave because of the decrease in its thickness and the higher localization of the flow. Thus, the effect of the reflected shock-wave diffraction in the region of transition on the mixture ignition should intensify with increase in the pressure, which is indirectly supported by the results of measurements of the induction time (Fig. 3).

However, the interaction of the reflected shock wave with the turbulent boundary layer does not completely explain such an unusual behavior of the temperature dependence of the induction time in the case where the density of the mixture increases. The experimental data should be analyzed with account for the time of this interaction after the reflection of the incident shock wave. If it is assumed that the critical Reynolds number in the region of transition to the turbulent boundary layer Re^* , calculated by the length l^* of the path of travel of gas along the wall downstream of the incident shock wave, is constant for the given setup, the length of the transition l^* , which is inversely proportional to the density downstream of the incident wave ρ_2 ($\sim Re^* / \rho_2$), will decrease with increase in the pressure of the

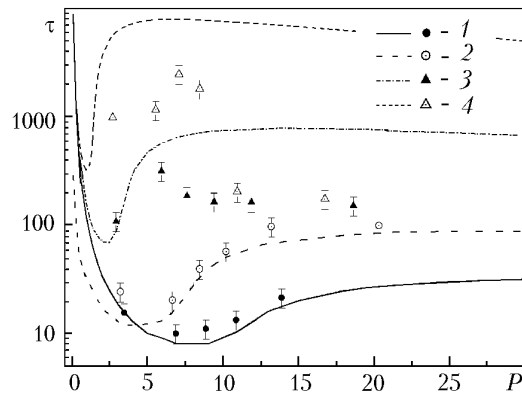


Fig. 4. Dependence of the induction time on the pressure calculated in accordance with the mechanism of [13] and measured at different temperatures of the hydrogen–air mixture downstream of the reflected shock wave: 1) 1260 ± 5 ; 2) 1200 ± 5 ; 3) 1090 ± 5 ; 4) 90 ± 5 K.

mixture and the region of transition will move to the face of the shock tube. This in turn will decrease the time interval between the instant the shock wave reflects and the instant it interacts with the region of transition. Therefore, the effect of the reflected wave bifurcation on the induction time of the mixture gradually increases with increase in the pressure (Fig. 3b and c) and practically completely determines the hydrogen ignition in the shock tube at a high density of the mixture, $\rho = 4.32 \pm 0.07 \text{ kg/m}^3$ (Fig. 3d).

Figure 4 shows calculated and empirical dependences of the induction time of the hydrogen–air mixture on the pressure at different temperatures of the gas downstream of the reflected shock wave. As is seen from the figure, the calculation in accordance with the mechanism of [13] at temperatures of $T > 1200$ K adequately reproduces the qualitative functional dependence of the experimental data for all the pressures studied. At the same time, it should be noted that the numerical simulation systematically gives ignition delay values that are 1.2–1.5 times lower than the analogous values given by the experiment, even though the inclusion of the effects of "nonideality" of the flow in a real setup should lead to the reverse result. Thus, the mechanism of A. A. Konnov (version 4.0) [13] and the GRIMECH 3.0 mechanism [15] should be somewhat refined to improve the quantitative agreement between the data obtained on their basis and the measurement data.

As the pressure increases and the temperature of the mixture decreases ($T < 1200$ K), the reflected shock-wave bifurcation begins to significantly influence the nonuniformity of the flow and the ignition of the hydrogen–air mixture downstream of the reflected shock wave. This leads to a marked decrease in the measured induction time as compared to the induction time calculated for the entirely uniform gas downstream of the reflected wave (Fig. 4). Under these conditions, any comparison of experimental and numerical data becomes very difficult and calls for knowledge of both the detailed kinetics of oxidation and the distribution of the local thermodynamic parameters in the three-dimensional structure of the flow downstream of the incident and reflected shock waves. At the same time, the effect of stabilization of the ignition delay and hydrogen combustion in the wide ranges of temperatures and pressures of the reacting mixture (Fig. 4, dependences 3 and 4) due to the interaction of the shock wave with the turbulent boundary layer is of considerable interest and should be taken into account when certain practical problems and applications are considered.

CONCLUSIONS

The ignition of a stoichiometric mixture of hydrogen with air at pressures of 2–21 atm and temperatures of 840–1430 K has been investigated numerically and experimentally.

It has been established that the kinetic mechanism of A. A. Konnov (version 4.0) [13] and the GRIMECH 3.0 [15] kinetic mechanism provide a good qualitative agreement of the data obtained on their basis with the experimental functional dependence of the hydrogen ignition delay on the pressure at temperatures of $T > 1200$ K; however, they should be somewhat refined to improve the quantitative agreement with the experimental data.

It has been shown that the interaction of the reflected shock wave with the boundary layer significantly influences the induction time measured and the regime of hydrogen ignition at mixture temperatures of $T < 1100\text{K}$. This effect intensifies with increase in the pressure and density of the gas downstream of the reflected shock wave and results in the dependence of the induction time on the temperature weakening and the ignition process stabilizing.

The data obtained can be used for analysis of the combustion process in engines and gas-turbine plants, improvement of the kinetic mechanism of the chemical reactions of hydrogen oxidation, and prediction of the limits of detonation and explosion safety of hydrogen–air mixtures under different conditions.

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NOTATION

f , focal distance of the lens, cm; l^* , length of the transition to the turbulent boundary layer downstream of the incident shock wave; P , pressure downstream of the reflected shock wave, atm; Re , Reynolds number; Re^* , critical Reynolds number; S , area of the observation region along the axis of the shock tube; T , temperature downstream of the reflected shock wave, K; t , time, μsec ; Δt , time interval, μsec ; U , intensity of the proper glow, V; ΔU , change in the proper glow intensity in a unit time; V_{32} , velocity of the detonation wave between the pressure transducers 2 and 3 of the measurement unit (Fig. 1); λ , light wave length, nm; $\Delta\lambda$, halfwidth of the transmission band, nm; ρ , density of the gas downstream of the reflected wave, kg/m^3 ; ρ^* , density of the mixture downstream of the reflected wave during the calibration, kg/m^3 ; τ , induction time, μsec .

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