

# Numerical Investigation of Hydrogen-Air Mixtures Ignition near Lean Flammability Limit

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## Abstract

The 0D and 1D approaches for lean flammability limit determination are presented for hydrogen-air mixture. Results, obtained with the use of two methods revealed the consistency of 1D approach rather than 0D. It is shown that the ignition of lean mixtures is unstable towards gasdynamic effects evolving on the scales of ignition zone. The other part of the paper investigates the concentration limits of flame propagation and demonstrates that the addition of the spatial domain with non-uniform concentration distribution to the uniform one could expand the concentration limits of flame propagation.

## Introduction

In the process of severe accident in nuclear power plant the breakdown of the reactor zone takes place. It causes a set of conditions for hydrogen release within the volume under the containment due to zirconium oxidation in fuel claddings. The convective flows formed under the containment redistribute hydrogen and mix it with air, vapor and other products of core melting and oxidation. As a result the containment volume fills with hydrogen-containing mixtures which carry a hazardous potential as they can be ignited by relatively low energy sources. Hereafter, the process could evolve in different ways: combustion quenches; combustion propagates through the combustible mixture as a slow or fast flame; accelerating flame transforms into the detonation; the detonation forms from the very beginning due to intense local energy release. All the outlined manifestations of the additional energy release in the combustion wave could be extremely destructive during a severe accident on a nuclear power plant. Obviously, the question of ignition criterion and combustion evolution under reactor containment is one of the fundamental questions in the assessment of hydrogen hazards. The complex criteria depend on a large set of particular conditions formed during the accident. It is exceedingly complicated to predict these conditions not only quantitatively but even qualitatively at least because majority of the accident details are unknown. On the other hand the uncertainty in formation of developed combustion is reflected on the reliability of prognostic results. In this case it is important to carry out the determination of gaseous mixture limit characteristics, beneath which the ignition of the mixture is not possible. Among such characteristics one can specify the lower limits of the hydrogen concentration in the mixture and the temperature at which the ignition takes place. The temperature as a criterion is less important in practice because in terms of severe accident the high temperature regions always emerge as a result of electric breakdowns or energy rejection from the overheated core melt. The composition of gaseous mixture can be rather complex, however at determining the concentration flammability limits it is sufficient to analyze pure hydrogen-air mixtures as some other

components like vapor or carbon dioxide could only provide the increase of the lower flammability limit.

The most rigorous criterion of the combustion initiation is the lower limit of exothermic reaction which has pure chemical origins and is determined by thermodynamic conditions providing the equality of chain branching and chain termination rates. One more obligatory criterion for combustion propagation through the combustible mixture is the equality of the ignition delay to the characteristic time of energy dissipation in the energy release zone. At the certain temperature and pressure this flammability limit depends on the concentration of fuel component. One should expect that as the burning-out of extremely lean mixtures is far from being complete and therefore represents the underdeveloped exothermic reaction with low heat release, this could not lead directly to the intense dynamic loads distorting the safety systems. However, the detailed analysis of flame propagation in hydrogen-air mixtures near lower flammability limit is immensely important as the "smoldering" sub-critical flame could originate the intensively accelerating flame in case of changing environment conditions (like additional hydrogen release into the burning area or propagation of the heat wave inside the region with higher hydrogen concentration).

One of the most constructive methods for analysis of transient ignition and combustion processes was proposed by Ya.B. Zeldovich in 1980 [1,2] when he solved both problems for non-uniform initial conditions applying the approach elaborated earlier by A.N. Kolmogorov, I.G. Petrovsky and N.S. Piskunov [3]. Such an approach allows to describe the combustion wave propagating along the non-uniformly heated or premixed medium as an "intermediate asymptotic". Different physical mechanisms "switch on" and become significant on the different time scales. Therefore, the process evolving along the non-uniformity should be determined by different physical mechanisms on the different stages of the process evolution. Thus if one takes the non-uniform distribution of the fuel concentration heated up to the same temperature the ignition will start independently in every part of the distribution. One should observe a so-called spontaneous combustion

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wave propagating along the non-uniformity (see e.g. [2]) until the chemical factors would play a greater role. In the region of the non-uniformity where the characteristic scales of chemical reaction become much larger than gasdynamic scales one should observe the switch between spontaneous and gasdynamic mechanisms of process evolution (the transport phenomena should play a greater role in further propagation of the wave front). The concentration at which such a switch takes place naturally determines the lower flammability criterion mentioned above. Thus such an approach allows estimating the lower flammability limit knowing the reliable features of the chemical kinetics. In case of the reaction wave propagating along the fuel concentration non-uniformity at normal conditions one should observe clear visualization of the criteria for combustion stability.

The present study reproduces the solutions of both the problems of ignition and flame propagation in the presence of hydrogen mass fraction non-uniform distribution. The solutions are obtained using direct numerical simulations involving gasdynamic model and detailed (reduced) kinetics of hydrogen-air combustion. The aim of the study is to get a new method for estimation of flammability limits and analysis of transient processes in the combustible mixtures of the lean-limit composition.

#### 0D method for LFL determination

Zero-dimensional combustion modelling, implying the solution of the system of chemical kinetics equations coupling with the equation for temperature reevaluation, represents the simplest method for LFL determination. Such a modelling enables to determine the induction time  $\tau_{ind}$  of the mixture at certain temperature, concentration and pressure, likewise the principal time of exothermic stage of combustion, which we will denote  $\tau_{reac}$ , at certain initial parameters as well.

In the present study, the induction time was determined as the time, passed from the starting point till the maximum concentration of H radicals achievement, and the  $\tau_{reac}$  time – as the time passed from the end of induction period till the condition at which the temperature achieves the stationary value. It is suffice to note that there is no universal criterion for the determination of the moment at which temperature achieves its final stationary value as distinct from the existing criterion for induction time. In such a case we chose our own criterion, which is not universal but good enough for determination of  $\tau_{reac}$  for the considered temperature and hydrogen-in-air concentration ranges. According to this criterion the time  $\tau_{reac}$  is considered to be achieved when the temperature variation at the next time step  $\Delta T$  becomes lower than the value of  $2.5 \cdot 10^{-4}$  K (when using time step of  $10^{-8}$  s).

From the comparison of induction time and  $\tau_{reac}$  time it is possible to distinguish two regimes of chemical transformation. At relatively low temperatures the leading role belongs to the mechanisms of chains termination and therefore the induction time is much

longer than the principal time of exothermic stage of combustion. At high temperatures the role of chain-branching reactions become significant and the induction time has a low value and falls short of  $\tau_{reac}$ .

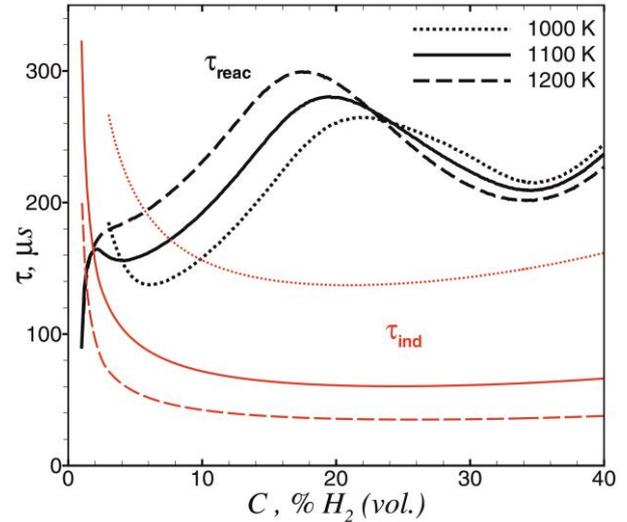


Figure 1. Induction time and principal time of exothermic stage of combustion of hydrogen-air mixture versus hydrogen concentration at different initial temperatures. Calculations were performed with the use of kinetics mechanism [4].

Transitional stage of chemical transformation between two regimes is characterized by the equality of  $\tau_{ind}$  and  $\tau_{reac}$ , and it is possible to determine conditions for its achievement: temperature (which is known also as the crossover temperature), hydrogen concentration and pressure. Since we assume the standard pressure (1.0 atm) throughout our investigation, the transitional stage is determined as a crossline of the surface defined by the  $\tau_{ind}(T,C)$  function and the surface defined by  $\tau_{reac}(T,C)$  in the space of  $(T,C)$ -coordinates. Such a set of transitional stages with corresponding values of temperature and hydrogen concentration could be regarded as a low flammability limit of hydrogen-air mixture at certain pressure as at lower temperatures and hydrogen concentrations the recombination (termination) reactions limit the ignition. In Fig. 1 dependences of induction time and  $\tau_{reac}$  time versus hydrogen concentration are presented for different ignition temperatures. The point of intersection of two curves corresponding to the same temperature value defines the so-called crossover stage, characterized by the temperature ( $T_{cr}$ ) and concentration ( $C_{cr}$ ) at which it is achieved. In Fig. 2 the values of temperature  $T_{cr}$  and concentration  $C_{cr}$  at crossover are presented. Calculations were performed with the use of different chemical kinetics mechanisms [4-8]. The figure shows that at sufficiently high temperatures the self-ignition of mixture is possible even if the concentration of hydrogen in mixture is approximately 1% (vol.) (concentration flammability limit). At relatively low temperatures the abrupt vertical course of any curve is observed, what demonstrates that almost at any hydrogen concentration the mixture could not be ignited if the temperature is less

than the value of 950 - 1050 K (for different kinetics mechanisms), nearby which the curves demonstrate asymptotic behavior (temperature flammability limit).

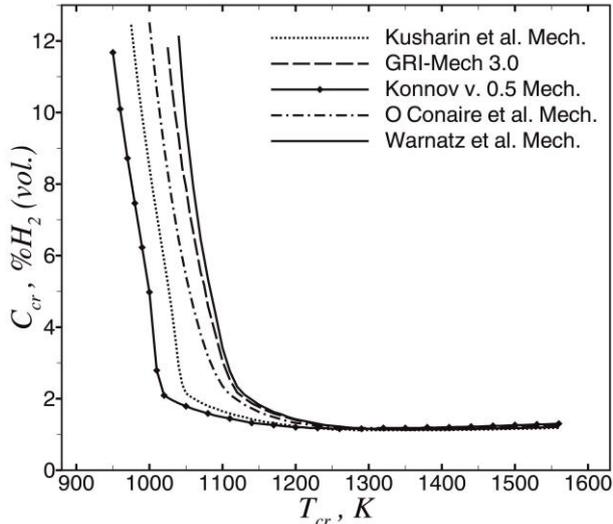


Figure 2. Temperature and hydrogen concentration, corresponding to crossover, obtained with the use of different kinetic mechanisms [4-8].

Thus, the LFL determined from 0D calculations turns out to be 1% (vol.) of hydrogen concentration (the value  $C_{cr}$ , which is approached by any curve in Fig. 2 at high temperatures). This result should be considered as underestimated comparing to the value of 4% (vol.), stated in numerous experiments [e.g., 9]. This criterion does not contain the information on how the ignition conditions affect the reaction evolution and competition between chain-branching and chain-termination mechanisms. In order to get more clear understanding of the mechanisms providing or preventing the ignition in lean hydrogen-air mixtures the 1D approach was elaborated that is presented in the next section. The kinetics scheme of Kusharin et al. [4] will be used in 1D experiments as it demonstrated average behavior for crossover curve among results obtained with the use of other schemes.

### LFL determination in 1D statement with concentration gradient

As it can be concluded from the previous section the ignition of rather lean hydrogen-air mixtures (1.5-2.0%) will become possible if the locally deposited energy is sufficiently large, what provides a high temperature inside the ignition zone. However the energy release in the exothermal reaction of the lean mixtures near the crossover limit is still rather small (e.g. it provides heat effect not greater than +240 K for 4% hydrogen-in-air mixture, Fig. 3). It is obvious that such a weak energy release cannot provide the self-sustained flame propagation, however it can be insufficient already on the ignition stage, when the gasdynamic effects evolves on the spatial scales of ignition zone. To understand the gasdynamic stability of the ignition in lean mixtures we have considered the following numerical setup. We studied a spontaneous wave of ignition propagating along

the linear gradient of hydrogen concentration. The setup is based on the concept of spontaneous combustion wave, which was considered by Ya.B. Zeldovich [2].

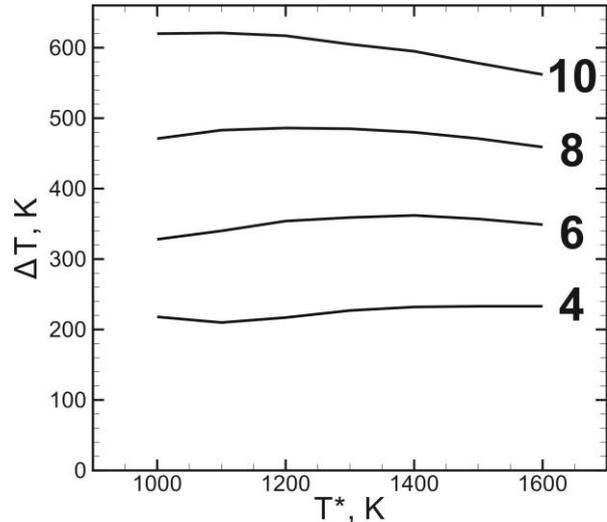


Figure 3. Temperature rise as a result of ignition of lean hydrogen-air mixtures at different hydrogen contents (designated by numbers) and different ignition temperatures ( $T^*$ ) (results were obtained in 0D simulations).

According to Zeldovich, if there exists a distribution of reactive mixture with an induction time gradient in some spatial domain, then the formation of spontaneous combustion wave is possible. This wave would spread along the gradient from the region with minimal induction time to the region with maximal one. The mechanism of such a wave is a consequence of independent explosions in different parts of gradient; therefore, the speed of the spontaneous wave is not limited by the gasdynamic velocity scales (like sonic speed). As the initially supersonic spontaneous wave propagates along the gradient inside the lean region its speed decreases with rising  $\tau_{ind}$ . The compression waves forming behind the propagating front of exothermal reaction overrun the front and the spontaneous wave stops existing. Further propagation of the temperature wave is caused by gasdynamic factors (convection, thermal conductivity, diffusion etc.). The transition between spontaneous and gasdynamic regimes of thermal wave propagation determines the point of ignition stability towards the external gasdynamic impact. In richer mixtures the kinetics is not limited by the gasdynamic factors, and in the leaner mixtures it is. Here we realized the described method and carried out a set of 1D calculations.

Numerical simulations used the mathematical model, representing the standard system of gasdynamics equations with account of heat transfer, mixture-averaged diffusion, viscosity and heat release due to chemical reactions. Problem statement was the following. At initial moment of time temperature and pressure (1.0 atm) were set uniform in 1D problem space domain. The initial temperature  $T_{initial}$  took the values

from the range of 950-1200K (typical for ignition conditions) for different simulations. The hydrogen concentration was set as a descending gradient: maximal concentration corresponded to the left-hand side of the domain, and its minimal value (zero concentration) – to the right-hand side. The value of maximal concentration was determined according to the stated initial temperature and induction time dependence on hydrogen concentration similar to that presented in Fig. 1. Thus, for 950 K it was equal to 16.2% (vol.), and for 1200 K – 25.8%. The size of 1D domain  $L$  was determined also with account of initial temperature and concentration as according to the formula:  $L = a_{max} \cdot (\tau_{ind})_{min}$ , where  $a_{max}$  – is a maximal sonic speed and  $(\tau_{ind})_{min}$  – is a minimal induction period on the left-hand side of the domain. This expression represents the condition of equality of maximal spontaneous wave speed and the maximal sonic speed in the considered problem setup. The choice of such a scale  $L$  excluded sufficiently supersonic regimes and allowed to extract more clearly the transitional point where the spontaneous wave stops its existing. For 950 K the length  $L$  was equal to 0.187 m, and for 1200 K – 0.027 m.

Hydrogen concentration profiles for sequential time moments with 10  $\mu$ s intervals are presented in Fig. 4. This figure shows that at some moment of time the autoignition of the mixture takes place nearby the left-hand edge, which is transformed into spontaneous combustion wave. This wave spreads in a direction of hydrogen concentration decrease.

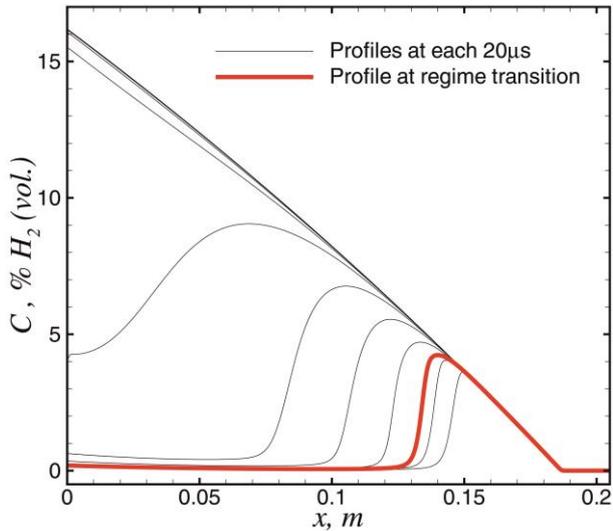


Figure 4.  $H_2$  concentration profiles through each 10  $\mu$ s from the initial time moment. The bolded profile corresponds to the moment of combustion regime transition. Simulation was performed at initial temperature 950 K.

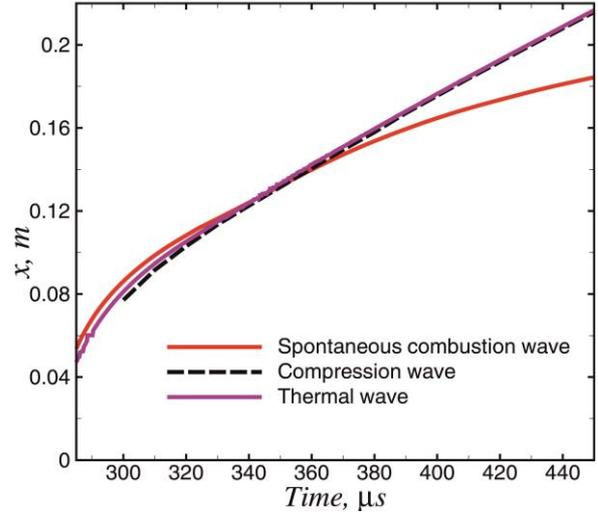


Figure 5. Trajectories of spontaneous combustion wave, thermal and compression waves. Simulation was performed at initial temperature 950 K.

When tracking the trajectories and velocities of combustion wave, thermal wave and compression wave it is possible to observe the existence of two distinct combustion regimes (Fig. 5). The position of spontaneous combustion wave front was tracked as a point with coordinate, at which hydrogen concentration spatial derivative takes its maximal value in absolute magnitude. In this way positions of thermal and compression waves were tracked, according to the maximal value of temperature spatial derivative and density local maximum correspondingly. Thermal wave formed at initial stage of combustion at a certain moment of time starts to be followed by compression wave, which originates due to formation of hot combustion products behind combustion front. Thermal and combustion waves spread jointly, being supported by heat release from combustion. At some moment of time, thermal wave being totally supported by compression wave and thermal conductivity begins to precede the decelerating spontaneous combustion wave. This moment of time marks the transition of combustion regime – the velocity of combustion front becomes nearly steady with subsequent falling below level of sonic speed. At that, the velocity of thermal wave remains to be supersonic during all period under consideration. Therefore, it may be concluded that the hydrogen concentration behind the combustion front becomes insufficient for thermal wave support. Determined with the use of concentration profile (Fig. 4) the value of  $H_2$  concentration at the moment of regime transition is equal to 4.2% (vol.) for simulation with initial temperature 950 K. In Fig. 6 the values obtained from simulations with different  $T_{initial}$  are presented. All determined values lay close to 4%, but as the temperature increases it drops to lower values. The latter denotes that higher intensities of energy source provide more stable ignition of the lean mixtures which is quite obvious.

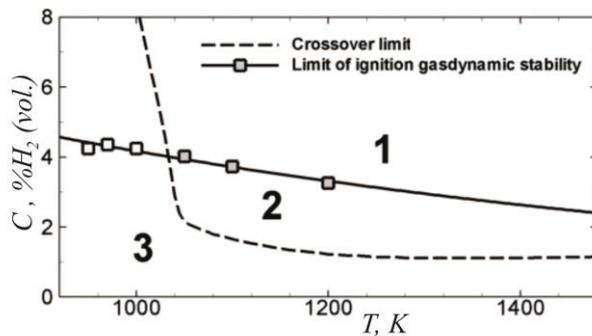


Figure 6. Limits of ignition stability for hydrogen-air mixture. 1 – the region of stable ignition, 2 – the region of gasdynamic instability of ignition, limited above by the curve obtained in 1D simulations (solid line with squares), 3 – the region of kinetic instability of ignition, limited above by the curve obtained in 0D simulations with use of  $\tau_{ind} = \tau_{reac}$  criterion.

### Determination of concentration limits for flame propagation

It is of great interest in addition to the obtained results to evaluate, at which hydrogen concentration in its mixture with air the formation of steady flame propagation is possible. Results of 0D calculations, presented in Fig. 2, showed that the initiation of exothermic reaction in hydrogen-air mixture is possible nearly at any hydrogen content. The temperature rises in the location of energy deposition as it is demonstrated in Fig. 3 and can be transferred by the diffusive and convective flows into adjacent regions.

The present simulations were performed for two different statements: with uniform and non-uniform distributions of hydrogen concentration in terms of ignition by high temperature spot. In the first case, the hydrogen concentration was taken equal to 4, 6, 8 and 10% (vol.) for different numerical experiments, the ignition temperature was equal to 1500K what according to the results presented in fig. 6 provides stable ignition of the considered compounds. The pressure was standard in every point of the space domain. Results of simulations, presented in Fig. 7 by dashed curves, show that at hydrogen concentration of 4 – 8% the wave of steady flame does not arise and combustion formed at initial stage distinguishes gradually or proceeds in non-steady manner with oscillations between flashing and damping (non-steady combustion was observed at 8 and 10%). At concentrations more than 10% the steady flame propagation takes place.

In the second statement we considered non-uniform distribution of concentration by adding the domain with linear hydrogen concentration increase up to 15% to the domain from the first statement. It is found (see Fig. 7) that in these conditions the thermal wave propagated out from the ignition zone is able to ignite the mixture in the richer region. Further combustion front propagation along the concentration gradient results in the formation of steady self-sustained combustion wave in the richer region.

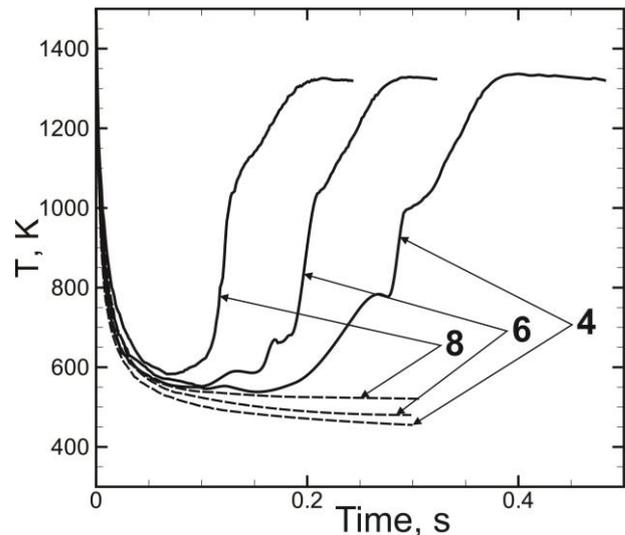


Figure 7. Evolution of temperature behind the diffusion wave front in case of uniform hydrogen concentration distribution (dashed curves) and behind the flame front (solid curves). Numbers designate corresponding hydrogen concentrations.

### Conclusions

In the present paper the 0D analysis as well as the 1D approach for lean flammability limit determination are presented. Results, obtained with the use of two methods revealed the consistency of 1D approach rather than 0D, what pays attention on the necessity of transport processes account for LFL determination. It is shown that the ignition of lean mixtures is unstable towards the gasdynamic effects evolving on the scales of ignition zone.

Besides, the concentration limits of flame propagation were evaluated. This set of numerical experiments showed that the steady flame does not arise if the hydrogen concentration is less than 10% in case of uniform concentration. However, in case of non-uniformly premixed medium the thermal effect of the lean mixture ignition can be sufficient to produce a thermal wave igniting the richer mixture in the adjacent region, where the self-sustained combustion is possible. Such a scenario should be taken into account while elaborating explosion safety systems.

### Acknowledgements

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