

# Numerical investigation of hydrogen-air mixtures ignition near lean flammability limit

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## 1 Introduction

Investigation of conditions for hydrogen-air ignition has an actual meaning in view of safety control on nuclear power stations. In the process of severe accident on nuclear power station the generation of hydrogen within containment could take place, what is caused by oxidation of fuel claddings. As hydrogen mixes with air the probability of hydrogen-air mixture ignition arises, taking into account the possible occurrence of high temperature spots during an accident. The ignition could evolve further in different ways: it could transit into detonation through combustion, or transit into stable/unstable combustion, or distinguish. In such a case in the light of safety the determination of limiting parameters, at which hydrogen-air mixture could not self-ignite at all, becomes crucial. These parameters are, firstly, temperature and hydrogen concentration. Since the temperature could be high enough due to electrical breakdowns during an accident the hydrogen concentration stands for the key parameter. The present paper aims to elaborate the correct method for determination of lean flammability limit (LFL) of hydrogen-air mixture at standard pressure and to analyze the probability of ignition transition into stable combustion in terms of uniform and non-uniform hydrogen concentration distribution near lean flammability limit.

## 2 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 from the end of induction period till the condition at which the temperature achieves the stationary value. It is suffice to note that the universal criterion for the determination of the moment at which temperature achieves its final stationary value could not exist 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  $\tau_{reac}$  in the targeted 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).

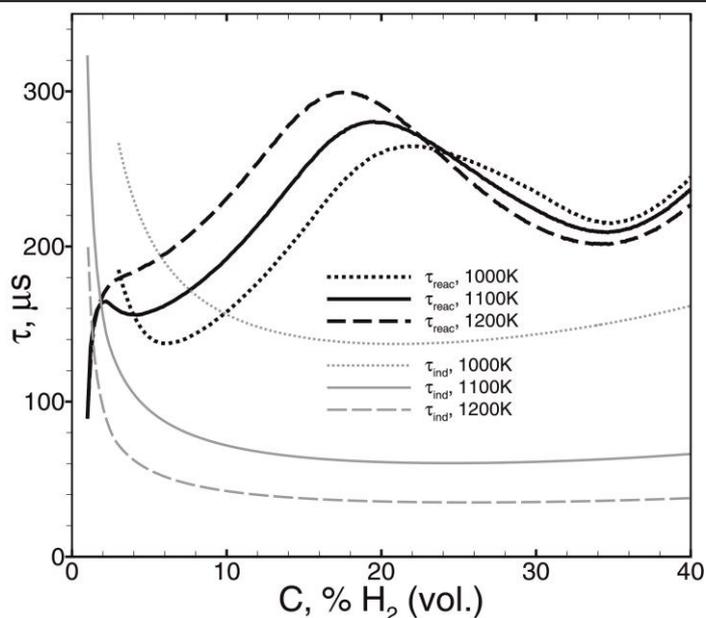


Figure 1. Induction time and principal time of exothermic stage of combustion of hydrogen-air mixture versus hydrogen concentration at different initial temperatures. Calculations were performed with the use of kinetics mechanism [1].

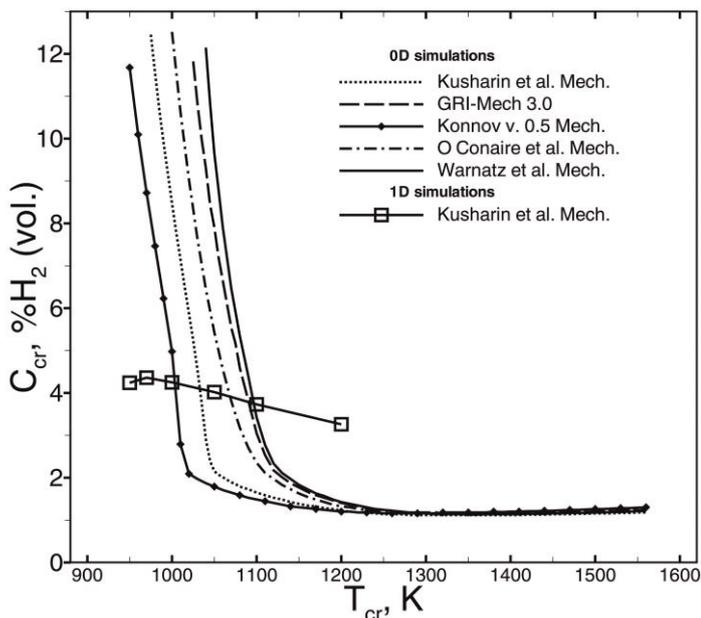


Figure 2. Temperature and hydrogen concentration, corresponding to crossover (designated as ‘0D simulations’). The values of LFL, determined in 1D calculations with concentration gradient statement (‘1D simulations’).

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 induction time is significant and is greater than the principal time of exothermic stage of combustion, while at high temperatures the induction time has a low value and falls short of  $\tau_{reac}$ . 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 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. In Fig. 1 dependences of induction time and  $\tau_{reac}$  time versus hydrogen concentration are presented at different temperatures. The point of intersection of two curves corresponding to the same temperature value defines the so-called crossover stage, characterized by the temperature and concentration 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 [1-5]. 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).

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., 6]. In order to make a correct determination of LFL we elaborated the specific 1D setup for numerical experiment, which is presented below. The kinetics scheme of Kusharin et al. [1] will be used in 1D experiments as it demonstrated average behavior for crossover curve among results obtained with the use of other schemes.

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

An approach for LFL determination elaborated in the present study is based on the concept of spontaneous combustion wave, which was considered earlier by Ya.B. Zeldovich [7]. 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. As an induction time gradient Zeldovich stated the temperature gradient, due to the unequivocal and monotonic dependence of one quantity on another. In the present paper aimed to determine LFL we assume the gradient of hydrogen concentration as an induction time gradient, what is justified by the monotonic dependence of induction time on hydrogen concentration (see Fig. 1).

Numerical simulations used the mathematical model, representing the standard system of hydrodynamics equations with account of heat transfer, mixture-averaged diffusion, viscosity and heat release due to chemical reactions. Problem statement was the following. At initial time temperature and pressure (1 atm) were set uniform in 1D problem space domain. The initial temperature  $T_{initial}$  took the values of 950 K, 970 K, 1000 K, 1050 K, 1100 K, 1200 K 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 according to the formula:  $L = a_{max} \cdot (\tau_{ind})_{min}$ , where speed of sound  $a$  and induction time  $\tau_{ind}$  were taken maximal and minimal correspondingly across the initial distribution in domain. This expression represents the condition for acoustic oscillations flattening in the domain of size  $L$ . For 950 K the length  $L$  was equal to 0.187 m, and for 1200 K – 0.027 m. The boundary conditions corresponded to the closed left-hand edge and to the opened right-hand one.

Hydrogen concentration profiles for sequential time moments at 10  $\mu$ s intervals are presented in Fig. 3. This figure shows that at some moment of time the autoignition of the mixture takes place nearby the

left-hand edge, which transforms into spontaneous combustion wave. This wave spreads in a direction of hydrogen concentration decrease.

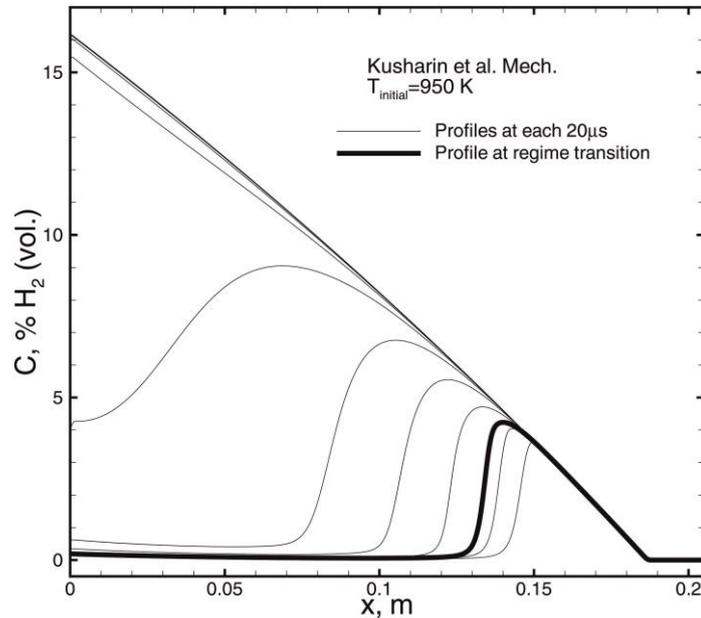


Figure 3.  $\text{H}_2$  concentration profiles through each 10  $\mu\text{s}$  from the initial time moment. The bolded profile corresponds to the moment of time at combustion regime transition. Simulation was performed at initial temperature 950 K.

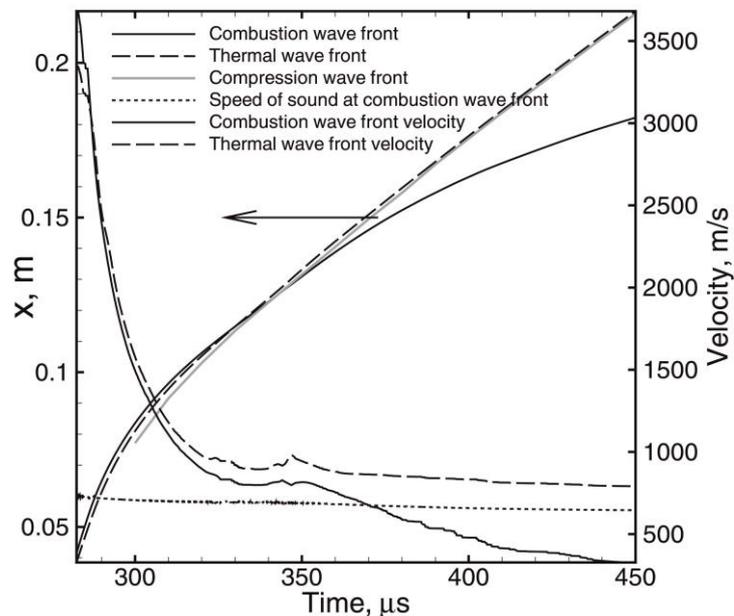


Figure 4. Trajectories and velocities of spontaneous combustion wave, thermal and compression waves. Sound speed at combustion front is presented. Simulation was performed at initial temperature 950 K.

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. 4). The position of spontaneous combustion wave front was tracked as a point with coordinate, at which hydrogen concentration derivative of  $x$  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 derivative

of  $x$  and maximal value of density 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 compression waves spread jointly, being supported by heat release from combustion. At some moment of time, thermal wave begins to precede the combustion wave, thereafter it is totally supported by compression 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 sound 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. 3) 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. 2 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 3.1%.

#### 4 Determination of concentration limits for flame propagation

It is of great interest in the present paper to evaluate, at which hydrogen concentration in its mixture with air the formation of steady flame propagation is possible. 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 temperature was equal to 1500 K and the pressure was standard in every point of the space domain. Results of simulations, presented in Fig. 5 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 was found (see Fig. 5) that even if the concentration in uniform region has a values of 4 – 8%, at some moment of time the formation of steady combustion occurs, what is originated by the thermal wave, arising in region with uniform hydrogen concentration.

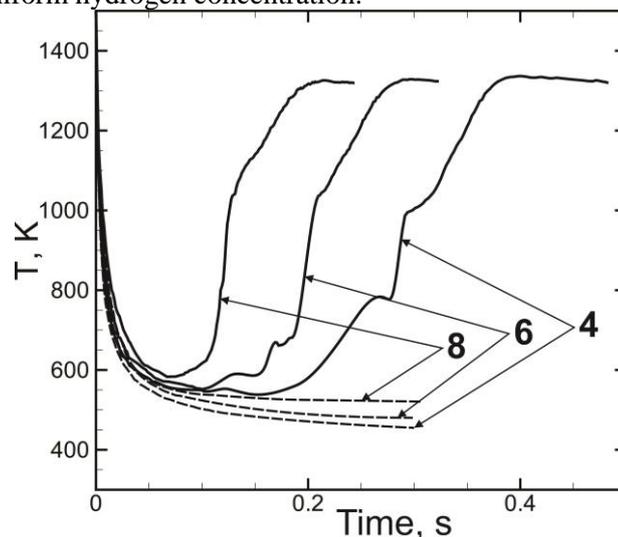


Figure 5. Evolution of temperature behind the diffusion wave front in case of uniform hydrogen concentration distribution (dashed curves) and behind the flame front (solid curves). Numbers designate corresponding hydrogen concentrations.

## 5 Conclusions

In the present paper the 0D analysis as well as 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. 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, at adding the domain with the concentration increase the steady flame exists even if the hydrogen concentration is less than 10% in uniform region.

## References

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