Peculiarities of mathematical modeling of combustion of hydrogen-air mixtures

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Abstract. Paper presents a comprehensive study of contemporary opportunities in the numerical analysis of transient combustion regimes on the example of the topical problem of hydrogen combustion. The elementary processes determining the development of combustion under different conditions are considered. In view of this analysis, it is concluded that the most appropriate for numerical modeling of classic deflagration inside confined space is the low-dissipation numerical schemes of low (second) order of approximation. For near-critical conditions where distinct mechanisms determine the combustion development one is able to utilize the simplifications such as low Mach number approximation.

1. Introduction

Nowadays the issues of hydrogen combustion attract a wide research interest that is related on the one hand to the hydrogen utilization as a perspective fuel and on the other hand to the hydrogen safety of industrial objects such as nuclear power plants. In the latter case, the abnormal or accidental scenarios developing on the industrial objects can lead to the generation, accumulation and subsequent explosion of hydrogen inside the confined space \cite{1}. When utilizing the hydrogen as a fuel for prospective engines \cite{2} the issues related to abnormal combustion of hydrogen are of importance since knock can take place and lead to the inoperability of the engine. In view of discussed applied problems, it is important to have a reliable technique for predicting possible combustion regimes arising on a wide range of spatial scales in a wide range of conditions. Such a technique should provide reliable predictions of flammability and combustibility limits \cite{3,4} on the basis of the contemporary theory of combustion and explosion. This paper analyses basic combustion regimes taking place in hydrogen-containing compounds and proposes numerical approaches that can be reliably utilized for numerical simulations of different combustion regimes. As a result, the hydrogen combustion efficiency and safety under certain conditions can be predicted with the application of such approaches.

Let us consider, as an example, the possible scenario of severe accident development on the nuclear power plant. Due to the destruction of the reactor zone the environment under containment is feeding with the hydrogen released from the reactor zone. Hydrogen mixes with air, water steam and other gaseous compounds generated in the destructed reactor zone, so the combustible mixture is forming \cite{1}. In such conditions, the combustible atmosphere inside
the congested volume of the containment building is forming, which can be ignited due to the energy release in the melted reactor zone. Close scenarios are possible also at the facilities for storage or transport of combustible gases for power engineering [5]. In both considered cases the simplest scenario can take place in which the explosion of a well-stirred hydrogen-based mixture evolves on relatively large scales (either inside confined space or explosive cloud). Exactly this particular scenario is one of the fundamental issues of the contemporary combustion theory and represents the basis for numerical approaches validation and verification.

In the case of near-stoichiometric mixtures, the deflagration wave can be formed which can be ignited even with the use of relatively weak energy source [6]. The mechanism of deflagrative combustion propagation is mainly related to the heat transfer and active radicals diffusion from the reaction zone into the adjacent layers of fresh mixtures. Moreover, the convective flows forming in the process of flame propagation affect the deflagration wave development. The crucial feature of the deflagration regime is that the hot combustion products expansion causes compression and motion of the fresh mixture. Inside closed volumes, this leads to the increase in pressure that is realized in the form of compression waves acting on the volume shell. In addition, these compression waves also determine the non-stationary dynamics of the flame itself, and one usually observes the temporal evolution of the explosion process [7]. The most hazardous scenarios of the deflagrative explosion are associated with significant flame acceleration and corresponding generation of shock waves or even onset of detonation [8].

When studying the combustion of ultra-lean hydrogen-air mixtures (with hydrogen content lower than 9%) no stable deflagration wave is observed. In such mixtures, a quasi-isobaric combustion regime establishes [9]. Herewith, the combustion proceeds in the form of so-called flame balls [10] affected by the buoyancy force and convective flows formed in the process of upward motion of the flameball [11]. The mechanism of such a combustion regime differs fundamentally from the classic deflagration regime. The combustion is mainly driven by the process of diffusion of deficient component (hydrogen) inside the hot flameball [12].

Both combustion regimes mentioned above are characterized by the development of numerous instabilities such as hydrodynamic instability [13], thermodiffusive instability [14], thermoacoustic instability, etc. Due to this to get a proper numerical representation of various combustion regimes when elaborating a mathematical model one should accurately take into account all the peculiarities of combustion wave evolution in particular conditions. In view of this, this paper is devoted to the detailed description of the combustion regimes arising in the hydrogen-air mixtures, mathematical models and numerical techniques for appropriate numerical representation of the regimes.

2. Mathematical models and numerical techniques
This section is devoted to the description of conventional mathematical models and numerical techniques traditionally used for numerical analysis of reactive flows gas-dynamics.

2.1. Compressible gas-dynamics of reactive flows
The conventional mathematical model for a wide range of problems related to the dynamics of reactive flows is based on the system of gas-dynamic equations with the account of compressibility, viscosity, thermal conductivity, multicomponent diffusion, and exothermic chemical reactions. The governing equations can be written in the following form:

\[ \frac{\partial \rho}{\partial t} + \frac{\partial (\rho u_i)}{\partial x_i} = 0 \]  \hspace{1cm} (1)

\[ \frac{\partial (\rho Y_k)}{\partial t} + \frac{\partial (\rho u_i Y_k)}{\partial x_i} = \frac{\partial (\rho Y_k V_{k,j})}{\partial x_i} + \rho \dot{\omega}_k \]  \hspace{1cm} (2)
\[
\frac{\partial (\rho u_j)}{\partial t} + \frac{\partial (\rho u_i u_j)}{\partial x_i} = \left[ \frac{\partial}{\partial x_i} (\sigma_{ji} - \delta_{ij} p) \right]
\]

(3)

\[
\frac{\partial (\rho E)}{\partial t} + \frac{\partial (\rho u_i E)}{\partial x_i} = \left[ \frac{\partial}{\partial x_i} (u_j \sigma_{ji} - u_i p) \right] + \frac{\partial}{\partial x_i} \left( \kappa(T) \frac{\partial T}{\partial x_i} \right) + \rho \sum_k \frac{h_k}{m_k} \left( \frac{\partial Y_k}{\partial t} \right)
\]

(4)

where \( x_i \) – spatial coordinates, \( t \) – time, \( \rho \) – density, \( Y_k = \rho_k/\rho \) – \( k \)-th species mass fraction, \( p \) – pressure, \( u_i \) – mass velocity vector, \( V_{k,i} \) – \( k \)-th species diffusion velocity vector, \( E = \varepsilon + \frac{1}{2} \left( \sum_i u_i^2 \right) \) – specific total energy, \( \varepsilon \) – specific inner energy, \( \dot{\omega}_k \) – chemical source term, \( T \) – temperature, \( \kappa(T) \) – thermal conductivity, \( \mu(T) \) – viscosity coefficient, \( h_k \) – enthalpy of formation of \( k \)-th species, \( m_k \) – \( k \)-th species molar mass.

Viscous stress tensor is given by:

\[
\sigma_{ij} = \mu(T) \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} - \frac{2}{3} \delta_{ij} \frac{\partial u_m}{\partial x_m} \right)
\]

(5)

To complement the governing system (1)-(4) equations of state in the following forms were utilized:

\[
p = \rho RT / \bar{M}
\]

(6)

\[
d\varepsilon = c_V(T)dT
\]

(7)

where \( R \) – universal gas constant, \( \bar{M} = \left( \sum_k Y_k/m_k \right)^{-1} \) – the average molar mass, \( c_V = \sum_k c_{V_k} Y_k \) – specific constant volume heat capacity of the mixture, \( c_{V_k} \) – specific constant volume heat capacity of \( k \)-th species.

2.2. Low Mach number approximation for near-isobaric flows

In case of quasi-isobaric conditions which, for example, can be realized in the process of ultra-lean combustion or in case of significantly large spatial scales where the compressibility effects become to play a negligible role it is useful to use a simplified system of equations describing the reactive flows in the approximation of low Mach numbers. This allows using the milder criteria for explicit numerical techniques which demand the choice of appropriate time step of integration.

\[
\frac{\partial \rho}{\partial t} + \frac{\partial (\rho u_i)}{\partial x_i} = 0,
\]

(8)

\[
\frac{\partial (\rho Y_k)}{\partial t} + \frac{\partial (\rho u_i Y_k)}{\partial x_i} = \frac{\partial (\rho Y_k V_{k,j})}{\partial x_i} + \rho \dot{\omega}_k
\]

(9)

\[
\frac{\partial (\rho u_j)}{\partial t} + \frac{\partial (\rho u_i u_j)}{\partial x_i} = \left[ \frac{\partial}{\partial x_i} (\sigma_{ji} - \delta_{ij} p) \right] + (\rho - \rho_0) g_i
\]

(10)

\[
\frac{\partial (\rho \varepsilon)}{\partial t} + \frac{\partial (\rho u_i \varepsilon)}{\partial x_i} = -p_0 \left( \frac{\partial u_i}{\partial x_i} \right)
\]

(11)

\[
+ \frac{\partial}{\partial x_i} \left( \kappa(T) \frac{\partial T}{\partial x_i} \right) + \rho \sum_k \frac{h_k}{m_k} \left( \frac{\partial Y_k}{\partial t} \right)
\]
Here $p'$ is the dynamic component of pressure fluctuations, which is by the order of magnitude much smaller compare with thermodynamic pressure $p_0$, which assumed to be constant in the process of ultra-lean flame propagation, $\vec{g} = (0, g)$ is the gravitational acceleration.

2.3. Conventional finite-difference schemes
One of the most commonly used approaches to simulate the dynamics of reactive gaseous mixtures is to use classical first- or second-order numerical schemes [15–17]. These schemes tend to introduce relatively high numerical viscosity in the solution, which could be slightly reduced only by utilizing extremely fine numerical grids. However, it is important to note that the stiffness of the problem related to the significant difference between the spatial and time scales on which the chemical and gas-dynamic processes evolve dictates the use of fine grids as well. Here it is useful to note that the characteristic spatial scale of the reaction zone is $\sim 0.1–1.0$ mm and can be by the orders of magnitude smaller than characteristic gas-dynamical scales of the problem. The close approach is to use high-order approximation schemes [18–21] that have less numerical viscosity. However, these schemes suffer from local accuracy degradation near discontinuities that can affect the solution significantly since transient combustion processes are characterized by complex gas-dynamical flows with various types of discontinuities [22, 23]. Another drawback of high-order schemes is the necessity of using flux limiting procedures to avoid high numerical dispersion [24], though the physical basis for such limiters could not always be justified and choosing optimal limiting procedure can be problem-dependent. Thereby utilization of the high-order CFD techniques for combustion modeling seems to be controversial compared to schemes with low accuracy orders.

2.4. Low-dissipation numerical techniques
Recent studies demonstrate the viability of contemporary second-order dissipation-free techniques for solving complex problems in a field of reactive gas dynamics [25–27]. The dissipation-free or low-dissipation numerical techniques, in particular, allows accurate enough representation of acoustics and complex gas-dynamic events involving the formation of vortexes, their interaction, their decay, etc. The dissipation-free CABARET method is a representative of this class of numerical schemes. It was fully developed and described in a series of papers by Golovizin et. al. [28,29]. CABARET method belongs to Godunov-type numerical method and possesses several unique features such as:

- the most compact computational stencil;
- second-order accuracy in space and time in problems with flows not requiring flow correction procedure;
- physical flow correction procedure based on direct usage of maximum principle for local Riemann invariants;
- possibility for acoustic waves propagation modeling against the background of strongly non-uniform in space and time flows;
- the numerical scheme does not cause attenuation or destruction of stationary vortexes, for propagating vortexes scheme dissipation effects are negligibly small;
- the numerical scheme is explicit and does not require any iterative procedures;
- in a number of cases absolutely dissipative-free and always has the least approximation dispersion.

The most disadvantage of the CABARET scheme is an operation with an extended set of flow parameters separated into two categories: “conservative” variables defined at cell centers and “flow” variables defined at cell faces. It imposes tight restrictions on computer random access memory capacities required for modeling via the CABARET method. However, the
resources of modern computational facilities easily meet these requirements. Above mentioned characteristics of the CABARET method stipulate the interest in possibilities of its appliance for solving unsteady reactive gas-dynamical problems.

3. Analysis of combustion regimes

3.1. Deflagration waves

Let first consider the classical regime of deflagrative combustion. After the deflagration front is forming due to localized ignition it propagates outwardly from the ignition zone with the initial speed of the order of laminar burning rate ($u_f$). Almost immediately the effect of combustion products expansion becomes notable that determines the flame acceleration up to the flame speed equal to $\theta u_f$ where $\theta = \rho_b / \rho_f$ is the so-called expansion ratio - the ratio of combustion products density ($\rho_b$) to the density of the fresh mixture ($\rho_f$). As it is known freely propagating flame is hydrodynamically unstable [13] that leads to its acceleration due to the development of so-called Landau-Darrieus instability on the flame surface [30]. It should be also noted that the non-steadiness of the combustion development on early stages including the transient manner of energy release [31], the stage of flame acceleration up to $U_{Fl} = \theta u_f$ and further flame acceleration due to the development of hydrodynamic instability determines the generation of the compression waves traveling out from the freely expanding flame front. Moreover, the energy release inside the thin reaction zone (inside the flame front) also produces the compression waves propagating out from the energy release zone. In view of this, it is of importance to reproduce well enough the patterns of acoustic fields generated by the expanding flame when modeling the process of deflagrative combustion. Of most importance are the cases of flame development inside the confined space. In such conditions, the acoustic waves irradiated by the expanding flame reflect from the walls and affect the flame front. In particular, this leads to the intensification of the flame front instability development via the mechanism of thermo-acoustic instability. As one can see from figure 1 a cellular structure develops on the flame surface that causes additional intensification of combustion and further flame acceleration.

As one can clearly see from figure 1 the numerically-reproduced cellular pattern on the flame front depends on the choice of numerical technique. Thus, the conventional numerical technique of the first order approximation provides a less pronounced cellular pattern with less spatial scales involved in the process. This is mainly due to the higher scheme viscosity which filters small-scale perturbation and makes the gas-dynamic patterns smoother.

The expansion of combustion products as well as the generation of acoustic waves determines also the compression of the gaseous mixture inside the confined space. Due to this the thermodynamic state of the fresh mixture changes in time that leads to the variation in burning rate and additional flame acceleration. Moreover, the compression waves generated by the expanding flame transfer the momentum to the fresh mixture and form directed flows determined by the geometry of the vessel. Thus, for example, when igniting the mixture near the closed end of the tube the flow directed out from the closed end towards the opposite end is forming [32, 33]. Such a flow possesses a peculiar structure characterized by almost constant velocity in the bulk flow and thin boundary layers near the tube wall. The interaction of the flame propagating through the tube with the established velocity profile causes the additional flame deformation and elongation along the tube wall. As a result, a so-called “tulip” flame is forming [34]. The interaction of the flame front with the flow is characterized by the positive feedback that determines the permanent acceleration of both the flame and the flow. At the same time the flame speed is not so high and much lesser than the sonic speed, so the compression waves transfer the momentum to the gas at a significant distance ahead of the propagating flame front. Due to this the boundary layer in the cross-section directly ahead of the flame front becomes thicker with time. Moreover, the boundary layer instability develops that leads to the formation of roller vortices [25, 26]. In such conditions, the velocity profile is changing with
time asymptotically aiming to become turbulent and cover the whole cross-section of the tube [35]. In view of this, it is important to reproduce with enough accuracy the development of boundary layers that can be achieved only when using the low-dissipation numerical techniques. Visualization of the flow in the vicinity of the flame front propagating in the tube is presented in figure 2.

As soon as the flame accelerates up to high enough speed the compression waves irradiated

Figure 1. Development of cellular front structure obtained via the CABARET approach. Solid lines represent isolines of temperature $T = 2800$ K in channels of 5 mm and 20 mm width.

Figure 2. Flow pattern ahead of the accelerating flame.
by the accelerated flame becomes to play an additional role in determining the formation of shock waves and even detonation [8]. At this stage, it also important to resolve fine enough the structure of the forming shock waves and reaction fronts. Herewith, it should be noted that at increased temperature and pressure the reaction fronts can become much thinner that makes it impossible to resolve them with enough accuracy with the same grid resolution as on the early stages of the process. When utilizing the low-dissipation numerical techniques it becomes much easier due to the fact that without a scheme dissipation the shock fronts are resolved with 1-2 numerical cells while the conventional low-order finite-difference schemes provide 5-6 numerical cells per shock front. So, when using the low-dissipative numerical techniques it becomes easier to reproduce the transient process of high-speed combustion even with the use of coarser numerical grids. In view of this, the calculation resources can be saved.

3.2. Flameballs in ultra-lean mixtures
As was mentioned above, the knowledge of near-critical combustion regimes is of paramount importance when estimating the critical conditions such as the lean flammability limit. On the other hand, the combustion regimes in such a near-critical region are characterized by significantly different physical mechanisms [11]. The combustion proceeds much slower than it does in the deflagration regime. Herewith, there is no significant pressure increase and moreover, the acoustic field generated by the propagating flame is negligible. Due to this, it is reasonable to utilize simplified mathematical models adopted for the analysis of incompressible or weakly compressible media. Such a numerical approach like one based on low Mach number approximation provides with high accuracy the flow patterns forming in the process of flameball upward propagation compared with the experimental data [36]. An example of flow visualization, in this case, is presented in figure 3.

![Figure 3. Evolution of ultra-lean hydrogen-air flame kernel rising due to the buoyancy force in terrestrial gravity conditions. An approximate flame lateral extension with time is indicated by arrows.](image-url)
4. Conclusions
Paper presents a comprehensive analysis of contemporary opportunities in the numerical analysis of transient combustion regimes on the example of a topical problem of hydrogen combustion. It is shown that the classic deflagration regime taking place in near-stoichiometric compounds is characterized by numerous non-steady processes associated with the gas-dynamic behavior of the flow inside the confined space. The process of non-steady deflagration development depends on the acoustic waves and vortices evolution in the environment. In view of this, the most convenient numerical instrument for analysis of such systems is the low-dissipation numerical schemes of low (second) order of approximation. When approaching the concentration flammability limits one faces the regimes of quasi-isobaric combustion which development is mainly related to the relatively slow gas-dynamic processes related mainly to the natural convection and to the buoyancy effect. In such conditions, it is convenient to utilize simplified approaches based on assumptions such as low Mach number approximation. It makes the process of numerical analysis easier and less demanding for the resources.

Acknowledgements
The research was partially funded by the Russian Foundation for Basic Research grant №18-38-20079. Yakovenko Ivan acknowledge financial support by the state support of young Russian scientists grant MK-3473.2019.2.

References
[1] International Atomic Energy Agency 2011 Mitigation of hydrogen hazards in severe accidents in nuclear power plants Preprint IAEA-TECDOC-1661 (Vienna: International Atomic Energy Agency)
[2] Ivanov M F, Kiverin A D, Smygalina A E and Zaichenko V M 2018 Technical Physics 63 148–151
[3] Coward H F and Jones G W 1952 Limits of flammability of gases and vapors Preprint Bulletin 503 (Washington, DC: Bureau of Mines)
[4] Shapiro Z M and Moffette T R 1957 Hydrogen flammability data and application to pwr loss-of-coolant accident Preprint WAPD–SC-545 (Pittsburgh, PA: Westinghouse Electric Corp.)
[5] Venetsanos A, Papanikolaou E, Delichatsios M, Garcia J, Hansen O, Heitsch M, Huser A, Jahn W, Jordan T, Lacome J M, Ledin H, Makarov D, Middha P, Studer E, Tchouvelev A, Teodorczyk A, Verbecke F and Van der Voort M 2009 International Journal of Hydrogen Energy 34 5912–5923
[6] Lewis B and von Elbe G 1959 Combustion, Flames and Explosions of Gases (New York: Academic Press, Inc.)
[7] Xiao H, Sun J and Chen P 2014 Journal of hazardous materials 268 132–9
[8] Kiverin A, Yakovenko I and Ivanov M 2016 International Journal of Hydrogen Energy 41 22465–22478
[9] Cashdollar K L, A Zlochower I, Green G M, Thomas R A and Hertzberg M 2000 Journal of Loss Prevention in the Process Industries 13 327–340
[10] Ronney P D 1990 Combust. Flame 82 1–14
[11] Yakovenko I S, Ivanov M F, Kiverin A D and Melnikova K S 2018 Int. J. Hydrogen Energy 43 1894–01
[12] Zel’dovich Ya B, Branblatt G, Librovich V B and Makhviladze G M 1985 The Mathematical Theory of Combustion and Explosions 1st ed (New York, NY: Consultants Bureau)
[13] Landau L D and Lifshitz E M 1959 Fluid Mechanics (Volume 6 of A Course of Theoretical Physics) (New York: Pergamon Press)
[14] Markstein G H 1964 Nonsteady flame propagation (New York: Pergamon Press)
[15] Smirnov N N, Nikitin V F, Stamov L I, Nerchenko V A and Tyrenkova V V 2017 International Journal of Computational Methods 14 1750038
[16] Kagan L and Sivashinsky G 2003 Combustion and Flame 134 389–397
[17] Ivanov M F, Kiverin A D, Klumov B A and Fortov V E 2014 Physics-Uspekhi 57 234–249
[18] Houmi R W, Ozgen A and Oran E S 2016 Combustion Theory and Modelling 20 1068–1087
[19] Valiev D, Bychkov V, Akkerman V, Law C K and Eriksson L E 2010 Combustion and Flame 157 1012–1021
[20] Xiao H, Houmi R W and Oran E S 2015 Combustion and Flame 162 4084–4101
[21] Lv Y and Ihme M 2014 Journal of Computational Physics 270 105–137
[22] Ciccarelli G and Dorofeev S 2008 Progress in Energy and Combustion Science 34 499–550
[23] Oppenheim A K and Soloukhin R I 1973 Annual Review of Fluid Mechanics 5 31–58
[24] Toro E 2009 Riemann Solvers and Numerical Methods for Fluid Dynamics A Practical Introduction (New York: Springer-Verlag Berlin Heidelberg)
[25] Kiverin A D and Yakovenko I S 2018 Physical Review Fluids 3 053201
[26] Kiverin A and Yakovenko I 2018 Physics Letters, Section A: General, Atomic and Solid State Physics 382 309–314
[27] Onuma Y 1993 Modeling of Turbulent Diffusion Flames (Tokyo: Springer Japan) pp 37–77
[28] Goloviznin V M and Samarskii A A 1998 Math. Model. 10 101–116
[29] Goloviznin V M and Samarskii A A 1998 Math. Model. 10 86–100
[30] Gostintsev Y A, Istratov A G and Shulenin Y V 1989 Combustion, Explosion, and Shock Waves 24 563–569
[31] Kiverin A D, Kassoy D R, Ivanov M F and Liberman M A 2013 Physical Review E 87 033015
[32] Ivanov M F, Kiverin A D and Liberman M A 2011 Physical review. E, Statistical, nonlinear, and soft matter physics 83 056313
[33] Ivanov M, Kiverin A, Yakovenko I and Liberman M 2013 International Journal of Hydrogen Energy 38 16427–16440
[34] Clanet C and Searby G 1996 Combustion and Flame 105 225–238
[35] Zeldovich Y B 1947 Journal of Technical Physics (in Russian) 17 3–26
[36] Volodin V V, Golub V V, Kiverin A D, Melnikova K S, Mikushkin A Y and Yakovenko I S 2019 Combustion and Explosion (Gorenie i Vzryv/(in Russian) 12 58–64