Extraction of Basic Flame Properties from Laminar Flame Speed Calculations

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Introduction

Development of reaction models for description of turbulent combustion and many practical applications related to safety analyses require data on basic flame properties, such as laminar burning velocity (S_L) and laminar flame thickness (δ), overall reaction order (n), effective activation energy (Ea). Not only data of unstretched flames are necessary, but also a response of the flame to straining, curvature and gas motions should be understood. A significant advance in theoretical description of these effects was made during last decades. The formulation of the corresponding models was based on the assumption of one-step overall reaction. Application of these analytical results and models in practice should require data on global flame parameters extracted either from experimental data or computational model of laminar flame with detailed reaction mechanism. In the present work, a problem of extraction of basic flame properties from data on computed laminar burning velocity is investigated. The objective is to develop a model for estimation of flame properties based on the Zeldovich description for the planar flame with one-step overall chemical reaction with high activation energy. Sun et. al. (1999) have demonstrated, that such an approach can give meaningful and useful results for hydrogen-air and propane-air flames. In the present work we develop this approach further, using as few simplifications as possible, comparing calculated and analytical flame properties. The problem of description of basic flame properties is investigated for hydrogen-air-steam mixtures at different initial temperatures and pressures. The choice of mixtures is connected with important practical applications in the field of nuclear reactor safety.

Analysis of existing methods for simulation of laminar burning velocities on the basis of detailed chemistry and real thermodynamic properties showed that existing, publicly available codes, such as Sandia Laboratories PREMIX and MIXFLAME, experience some problems with calculations for the mixtures close to limits of propagation. In the present work, a newly developed computer code for laminar burning velocity calculations is used, which is able to calculate very lean and reach mixtures important for practical applications. The computer code was validated first against data of specially conducted tests for a wide range of hydrogen-air-steam mixtures.

Experimental

The spherical bomb method proposed by Lewis and Elbe (1934) and developed by Andrews and Bradley (1972) was used for the experimental measurements in this work. The main and simplest technique in this method is to measure pressure rise in time at initial stage of flame propagation. The problem is that the fine pressure variation is to be measured very precisely in relatively short time. Reliable technique to do that has been available only recently. That is why this method is becoming very attractive because of its simplicity in equipment and treating the experimental data. Special provisions were made to avoid influence of the ignition source at early stage of mixture combustion. The use of thyratron switch allowed to shorten the spark duration to 5 μs and provide a solitary spark pulse with no oscillations. Precise measurements of small pressure variations provide an accuracy of about 10 % of the obtained burning velocity values. In addition, no significant difficulties were found to extend the technique for tests at elevated temperature and pressure. The mixtures were tested in a wide range of compositions; equivalence ratio varied from 0.2 to 5.5, temperature from 293 to 473K and pressure from 1 to 2 bar. The velocity values measured ranges from 0.033 to 5.92 m/s.

Model

The earliest efforts of solving premixed flame problems with multi step chemical kinetics were made with the use of shooting technique. However, if kinetics becomes more complex these methods are unusable. It was first introduced by Spalding (1956), that the use of implicit finite difference method, obtaining the solution of the steady-state problem as the asymptotic limit of a transient problem could be used for the flame calculations, further Warnatz, Wilde and others extended this method.

In the present work, for the solution of the nonstationary conservation equations describing freely propagating laminar flames, the finite difference method with a parabolic approach for three neighboring grid points is used. The calculations are carried out with the use of additional packages. For a chemical reaction rates and thermodynamic properties calculations the CHEMKIN package is used, and for transport properties – the TranFit package.

Calculations for hydrogen-air-steam mixtures were carried out with the use of chemical kinetic data from Gas Research Institute (GRI-Mech 2.11). The calculated laminar burning velocity values compared with experimental data are presented as an example in Fig. 1.

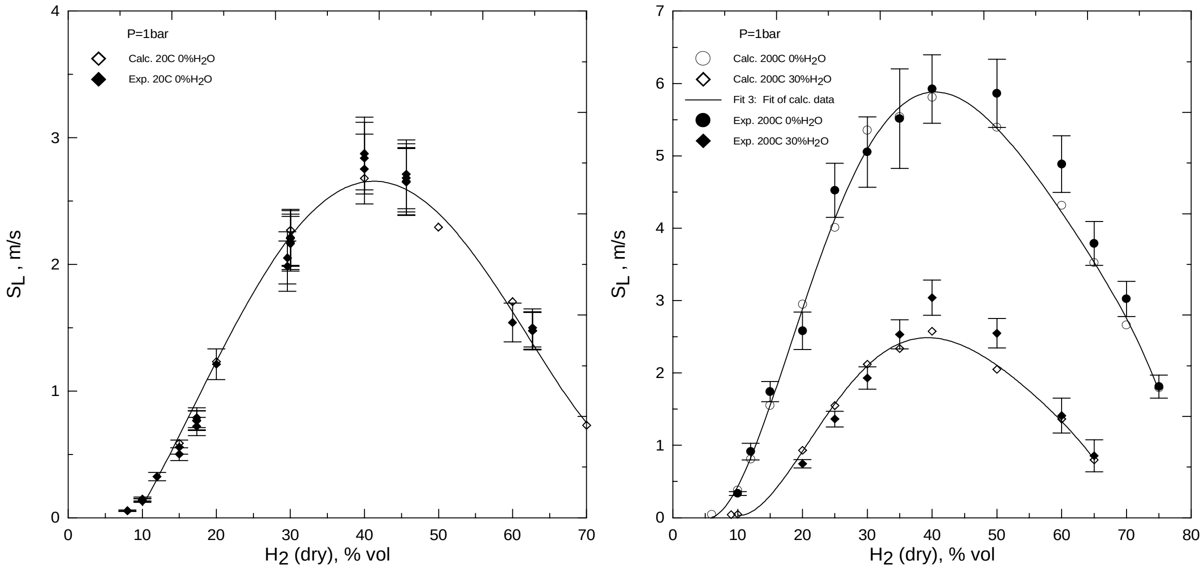

Figure 1. Laminar burning velocity values for hydrogen-air-H_2O mixtures at 20 °C, 0% (vol.) H_2O and initial pressure 1 bar versus hydrogen (dry) concentration (left figure) and hydrogen-air-H_2O mixtures at 200 °C (right figure). Comparison between calculations and experiment. Lines are polynominal fit through calculated points (empty symbols).

Extraction of basic flame properties

Extraction of basic flame properties is based on well-known equation authentic for laminar flame derived in the form (1):

$$\sigma S_L = \left(k_0 e^{-\frac{E_a}{T_b}} \frac{2\nu X_1^{n_1-1} X_2^{n_2} \left(\frac{p}{p_0}\right)^{n-1} W \chi\left(T_b\right) Le^{n} T_b^{2(n+1)} \Gamma_{n+1}}{\left(T_b - T_u\right)^{n+1} \left(\frac{T_u}{T_0}\right)^{n-1} E_a^{n+1}} \right)^{\frac{1}{2}}, \qquad (1)$$

where E_a is overall activation energy, T_b, T_u -burning and initial temperature, χ -thermal diffusivity, ν -stoichiometric coefficient, X_1, X_2, n_1, n_2 – volume fractions and reaction orders of limiting and abundant components, Le- Lewis number, W –mean molecular weight, σ -expansion ratio, S_L –laminar burning velocity, Γ_{n+1} –gamma function and k_0 –pre-exponential factor, $n=n_1+n_2$ –overall reaction order. The overall activation energy E_a represents the sensitivity of the global chemical reaction rate to flame temperature variations, and overall reaction order n indicates the pressure effect on the burning intensity.

With a small variation of initial pressure p, temperature T_b and expansion ratio σ are nearly constant, and one can obtain the following equation for overall reaction order:

$$n = 2\frac{\partial \ln(S_L)}{\partial \ln(p)} + 2\,. \tag{2}$$

A simplified equation for extraction of effective activation energy proposed by Egolfopoulos and Law (1990) is based on a simple heat and mass balance, and retains only the Arrhenius temperature function as the strongest one. This approach yields:

$$E_a = -2\left(\frac{\partial \ln(\rho_0 S_L)}{\partial (1/T_b)}\right)_p\,. \tag{3}$$

In terms of Eq. (1) this means that variations of S_L with T_b are prescribed to the Arrhenius term only. Generally, it is not necessary to use this simplification. Retaining all terms in Eq. (1) and assuming that k_0, E_a, n and Le are constant under little pressure and inert diluent variation, derivation for the overall activation energy can be written as follows:

$$E_a = \frac{\Delta\left[(2n+2)\ln T_b + (n-1)\ln p + \ln(W) + (n_1-1)\ln(X_1) + n_2\ln(X_2) - (n+1)\ln(T_b - T_u) + \ln(\chi) - 2\ln(\sigma S_L)\right]}{\Delta\left(\frac{1}{T_b}\right)}\,. \tag{4}$$

For E_a calculations, variations of T_b can be achieved adding or removing a small portion of inert species (e. g., N_2). This can be made keeping the pressure constant, or allowing corresponding pressure variations. In the first case one can extract E_a using both Eqs. (3) and (4), otherwise Eq. (4) should be used.

Examples of overall reaction order and effective activation energy calculations are shown in Fig. 2. Values of activation energy calculated with Eq. (4) and calculated from mass burning rate (Eq. 3) are quite different, especially near stoichiometry. This difference shows that effective activation energy is very sensitive to simplifications used to extract this parameter from laminar flame speed variations.

Another important flame parameter, the thermal thickness of laminar flame, δ, can be directly evaluated from the calculated temperature profile by using the gradient definition:

$$\delta = \frac{(T_b - T_u)}{\left(dT/dx\right)_{\max}}\,. \tag{5}$$

An example of flame thickness δ evaluation is shown in Fig. 3.

To extract Lewis number the approach introduced by Sun et. al. (1999) can be used. It is based on integral analysis of stretched flames. The main equation of this analysis describes the difference in burning velocities of stretched (S_L) and unstretched flames (S_L^0):

$$S_L = S_L^0 - \left[\frac{\sigma(\alpha-1)\beta(1-Le)}{2(1-\sigma)} + \frac{1-\alpha}{1-\sigma} - 1 + \sigma\right]\frac{2\dot{R}\delta}{R}\,, \tag{6}$$

where $\beta=E_a(T_b-T_u)/(RT_b^2)$- Zeldovich number and $\alpha=1+\ln[\sigma+(1-\sigma)e^{-1}]$ is the factor accounting for the thermal expansion effect.

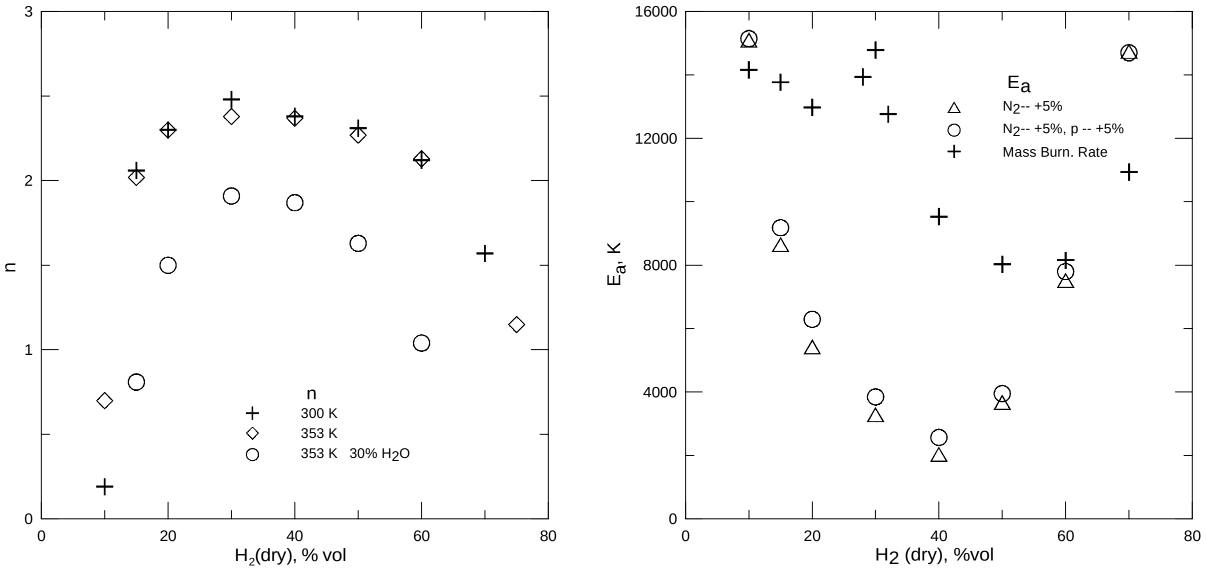

Figure 2. Overall reaction order *n* for hydrogen-air-H_2O mixtures at different temperatures and initial pressure 1 bar (left figure). Overall activation energy E_a for hydrogen-air mixtures at 300K and initial pressure 1 bar. (right figure).

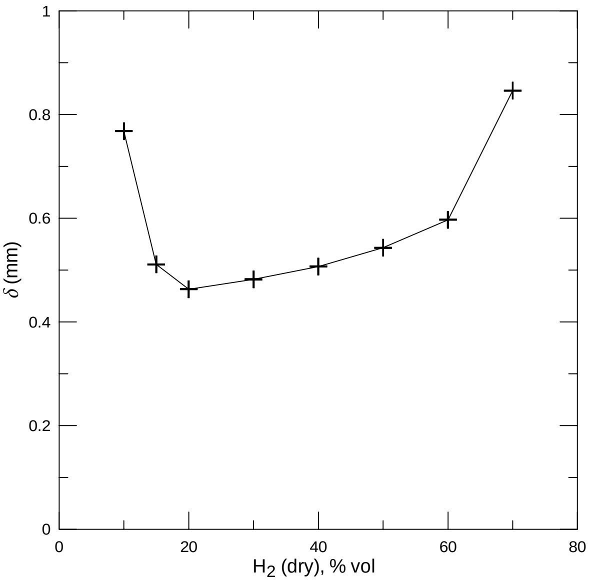

Figure 3. Laminar flame thickness δ for hydrogen-air mixtures at 300K and initial pressure 1 bar.

According to Eq. (6), we define Lewis number as:

$$Le = \frac{\Delta\left[\left(S_L^0 - S_L\right)\frac{R}{2\dot{R}\,\delta} - \frac{1-\alpha}{1-\sigma} - \left(\sigma - 1\right)\right]}{\Delta\left[\frac{\beta\sigma\left(1-\alpha\right)}{2\left(1-\sigma\right)}\right]}. \tag{7}$$

So, to extract *Le* one needs to calculate flame thickness and laminar burning velocity for stretched and unstretched cases and for two different pressures (*Le* is insensitive to pressure).

For very lean and rich mixtures *Le* can be easily evaluated as a ratio of thermal to mass diffusivity of limiting specie H_2 or O_2, and equals 0.33 and 2.32 respectively. Lewis numbers calculated with Eq. (7) are in a good agreement with these limits for very lean and reach mixtures. An application of Eq. (7) for extraction of Le is limited to the cases when $\beta >> 1$. This makes problematic to use this approach for a certain range of mixture compositions. For example, nearly stoichiometric hydrogen air mixtures (see Fig. 2) have low values of β, and calculations with Eqs (7) and (8) cannot give meaningful results.

Summary

The computer code for laminar burning velocity simulation able to compute mixtures close to the limits of propagation was developed. The comparison between calculated and experimental data on laminar burning velocities shows rather good agreement for hydrogen-air-steam mixtures for normal and elevated initial temperature and pressure.

The problem of extraction of basic flame properties from data on computed laminar burning velocity was studied. The Zeldovich equation for laminar burning velocity with one-step global reaction is used with all essential terms for extraction of flame properties. It was found that effective activation energy is very sensitive to simplifications used to extract this parameter from laminar flame speed variations.

The developed computer code and methodology can be used to extract overall effective activation energy (E_a), overall reaction order (n), laminar flame thickness (δ) from mixture composition and initial conditions for hydrogen-air-steam mixtures. The extracted basic flame properties can be used then for analysis of hydrogen combustion behavior important for nuclear reactor safety.

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