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Time-Dependent Solution of Pre-Mixed Laminar Flames with a Known Temperature Profile

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A computer program designed for the evaluation of molecular flows interacting through chemical kinetics and molecular diffusion is described. Measured values of temperature profile and mass flow are used. The starting profiles and the hot boundary values are calculated by a kinetics approximation found by neglecting diffusion. A time-dependent method is used together with successive grid refinements. The successive grid refinements reduced the execution times by a factor of 5 for a H_2/air flame at a pressure of 1 atm. For a CH_4/O_2 flame at 0.05 atm the reduction due to grid refinements was a factor 50 or more according to the estimations. The execution times for the test flames were a factor 4 slower than a current implementation of the steady state method. Possible optimizations of the present time-dependent version can decrease that difference significantly. The computed concentration profiles agreed with published computed results within 1%.

1. INTRODUCTION

Chemical physical systems consisting of many molecules and interacting through microscopic and macroscopic processes are of great importance in nature. Such systems can be found in, for example, atmospheric chemisty [1], combustion [2], and in general chemical kinetics [3]. Detailed computer modeling improves understanding of these complex chemical physical systems.

We have written a computer program to be able to analyze stationary flows of fluids with molecules interacting through chemical kinetics and molecular diffusion. The values of pressure and mass flow are assumed to be known from measurements. The same also applies to the temperature profile along the fluid stream. Chemical systems suitable for modeling by our program are, for example, thermal reactors and pre-mixed laminar flames (flat flames). Flat flames are excellent tools for studying important parts of complex combustion processes in, for example, engines [4]. Below we restrict our discussion of the computer program to

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flat flame applications. Our short term objective for writing the computer program was to analyze experimental concentration profiles from premixed laminar methanol/air flames [5].

Many people have contributed to flame computer modeling since the start in 1953 by Hirchfelder *et al.* [6]. As far as we know, only the computer program documented by Smooke [7] has been specially designed for evaluating experimental flat flames (but see also [8]). In that program a steady state method was used (for earlier references consult [9–11]). We use the time-dependent method which is regarded as less efficient than the steady state method. But the time-dependent method can, contrary to the steady state method, also describe processes such as ignition and relaxation phenomena. Since the introduction of Spalding in 1956 [12] many different versions of the time-dependent method have been presented (see, for example, [13–24]).

Recently Smooke [6] stated that the steady state method is 5–10 times faster than the conventional time-dependent method. Such a difference is tolerable for one-dimensional calculations, but could be intolerable for two- and three-dimensional calculations. In the work of validating our program we compared it thoroughly with the steady state program documented by Smooke [7]. Previously we have also made a comparison [5] with results obtained from the time-dependent program by Lund [19].

A major objective is to use physical and mathematical models (often well known) at an accuracy level of present flame experiments. Our implementation of a flame program can be summarized as follows: (i) a kinetics approximation gives the starting profiles, (ii) a simplified physical and mathematical model of diffusion is given, as compared to [7], (iii) the ordinary space coordinate is used all the time with second order central differencing, except for the mass flow term which is differentiated by the upwind method, (iv) the ordinary differential equations are solved with a first order implicit method and with a Jacobian formed analytically, (v) the resulting equation system is solved with a linear least square solution method, and (vi) successive grid refinements [7] are used to decrease execution time.

A main point in this study is the use of successive grid refinements to reduce the execution time. In the recent, very successful, version of the steady state method successive grid refinements have been used primarily to secure convergence [7]. Previously grid refinements have been used in computational physics by various authors as stated in [7] (see, for example, [25–28]). Nobody seems to have used (or proposed) successive grid refinements in time-dependent flame studies before now.

2. PROBLEM FORMULATION

We concentrate on simplifying the physical model and thereby reducing the mathematical problems as well as the computer cost. Only physical effects which are significant in relation to the experimental uncertainties in flame studies are introduced. Naturally, this means that the content of the model is simple and often well known.

2.1. *The Physical Background of Flat Flames*

A flat flame can be seen as a non-isothermal wall-less reactor. In our physical model of the flat flame we make the normal assumptions (idealizations). The gaseous fuel is mixed with oxidizer and inert gases in a prechamber. Ideal gas behavior is assumed. The pre-mixed gas flows with a constant mass flow through a wide porous burner. A laminar flow is induced in the emerging gas. After ignition a steady flame can be stabilized for certain combinations of parameters. The steady flame burns at a constant pressure. No temperature or concentration gradients exist parallel to the burner; i.e., the flame is one-dimensional. The burning gas mixture behaves as a continuous fluid. These are the conventional assumptions used in flame modelling. A thorough discussion of these and other flame properties can be found in the monograph by Fristrom and Westenberg [29].

2.2. *Mathematical and Numerical Description of the Flat Flame*

Conservation equations. The continuity equation for the one-dimensional flame is

$$\frac{\partial \rho}{\partial t}+\frac{\partial(\rho v)}{\partial z}=0 \tag{1}$$

with the corresponding equations for the species

$$\frac{\partial X_i}{\partial t}=\left(R_i-(\rho v)\frac{\partial X_i}{\partial z}-\frac{\partial(\rho X_i V_i)}{\partial z}\right)\Big/\rho \tag{2}$$

or

$$\frac{\partial X_i}{\partial t}=\text{chemical kinetics}+\text{mass flow}+\text{diffusion}, \tag{3}$$

where

X_i = the mole fraction per unit mass ($\sum X_i M_i = 1$)
t = the time (s)
R_i = the rate of production of species i by chemical reactions (mol/cm^3, s)
ρ = the density (g/cm^3)
v = the mass-averaged velocity of the gas mixture (cm/s)
z = the height above the burner (cm)
V_i = the diffusion velocity of species i (cm/s).

The computer program computes the concentrations as a function of the distance from the burner for the flame species. The conservation equation for energy is not used in the program, since we designed the program for evaluating experiments with known temperature profiles. Instead we used the measured temperature versus distance and the measured mass flow as input parameters. This approach is almost always necessary due to the cooling of the flame by the burner [7].

Chemical kinetics. An elementary reaction consists of at most three participants. Therefore we can write for a general reaction pair, composed of a forward and a backward elementary reaction,

$$S_i + S_j + S_k = S_l + S_m + S_n, \tag{4}$$

where $S_i, S_j, \ldots, S_n$ are the reacting species. If, for example, two identical species recombine we write that species number twice. For reactions containing fewer than three participants, the reactions are complemented with fictive participants at a concentration of unity, to simplify the numerical evaluation.

The rate of production from this general reaction pair is

$$r = k_f[S_i][S_j][S_k] - k_r[S_l][S_m][S_n] \tag{5}$$

with the kinetics contribution to the conservation equations from the pair of reactions as

$$r_i = r_j = r_k = -r \tag{6a}$$

$$r_l = r_m = r_n = +r. \tag{6b}$$

The total kinetics contribution can then be obtained by summing over all reactions. The partial derivatives in the Jacobian can be obtained from Eqs. (5) and (6). For the rate constants k_f and k_r a modified Arrhenius temperature-dependence is assumed:

$$k = AT^B e^{-E/RT}. \tag{7}$$

The coefficients for the forward rate constants can be taken from published data of the reaction mechanism; the coefficients for the reverse rate constants can be computed from the forward rate expression and a similar expression for the equilibrium constants

$$k_r = k_f/K. \tag{8}$$

Mass flow. For the mass flow term

$$(\rho v)\frac{\partial X_i}{\partial z}\bigg/\rho \tag{9}$$

the constant (ρv) is taken from the experimental evaluation at the cold boundary at burner $z=0$. The derivative is numerically calculated by upwind differencing at the grid points z_v using

$$\frac{\partial X_i}{\partial z}=(X_{i,v}-X_{i,v-1})/(z_v-z_{v-1}). \tag{10}$$

Diffusion. In the hydrocarbon–air systems, which are our main interests, the nitrogen dominates the gas mixture and we can assume that each species diffuses approximately as traces in nitrogen. From previous experience of flames [29] and flame modeling [30, 19, 23] it can be expected that a such a model would be accurate for many cases. The diffusion velocity V_i is therefore approximated [29] by

$$V_i=-\frac{D_{ij}}{X_i}\frac{\partial X_i}{\partial z} \tag{11}$$

where the index j stands for the nitrogen gas ([29], see also [30]). Naturally it also applies equally well with other dominant inert substances, such as argon, or systems with a large excess of oxygen. In the numerical evaluation of the diffusion term at a certain grid point, second order central differencing was applied.

The diffusion coefficients D_{ij} are computed from

$$D_{ij}=1.66\times 10^{-3}\,(M_i^{-1}+M_j^{-1})^{1/2}T^{1.67}/(\rho\sigma_{ij}^2(\varepsilon_{ij}/k)^{0.17}). \tag{12}$$

σ_{ij} and ε_{ij} are the Lennard–Jones $12-6$ potential parameters. The error in that description is about 2% and small in relation to the uncertainties in the Lennard–Jones $12-6$ parameters [29]. We calculate σ_{ij} and ε_{ij} from σ_{ii} and ε_{ii} and the simple combination rules $\sigma_{ij}=(\sigma_{ii}+\sigma_{jj})/2$ and $\varepsilon_{ij}=(\varepsilon_{ii}*\varepsilon_{jj})\;1/2$. For molecules we used values for σ_{ii} and ε_{ii} from viscosity measurements given by Fristrom and Westenberg [29]. The values for the atoms O and H were taken from Warnatz [22]. For radical species such as OH, HO_2, and HCO no literature values of σ_{ii} and ε_{ii} exist. Instead we calculated the effective diffusion diameter $(p\sigma_{ij}^2(\varepsilon_{ij}/k)^{0.17})$ for these species by an interpolation of the known effective diameters for similar molecules.

3. METHOD OF SOLUTION

3.1. *Kinetic Starting Approximation and Boundary Conditions*

The flame conservation equations for species must be solved by iteration using starting profiles for the species. The execution time necessary is assumed to depend strongly on the initial approximation [19, 31].

A steady flame can be looked on as a chemical kinetics system smoothed by diffusion. For the steady state equation system

$$0=R_i-(\rho v)\frac{\partial X_i}{\partial z}-\frac{\partial(\rho X_i V_i)}{\partial z} \tag{13}$$

an approximate solution can be found by setting the diffusion contribution to zero:

$$\frac{\partial X_i}{\partial z} = R_i(T(z))/(\rho v). \tag{14}$$

From measurements we take the concentrations of fuel, oxidant, and inert substances and the total mass flow at the cold boundary. Here as in the time-dependent calculations, with diffusion included, the measured temperature profile $T(z)$ is also used. This way the approximation gives a first order description of the specific flow-time scale, thermal conductivity, and diffusion. Previously Ay and Sichel [32] described a flat flame qualitatively by using a conventional kinetics calculation started at 1000 K and with a temperature evolution. Unfortunately their approach gave a wrong length-scale (flow-time scale). The flow-time, as seen by a fluid particle traveling upwards, is calculated from the expression

$$(\rho v)_{z=0} = \rho \frac{\Delta z}{\Delta t}. \tag{15}$$

The flow-time is a characteristic time for the combustion system studied.

In the solution procedure of the kinetics approximation a flow-time scale, calculated from Eq. (15), is used instead of the distance scale. At each flow-time step a chemical kinetics mean gradient is calculated. This kinetics gradient and the graphs of the species profiles give information about where the hot boundary can be placed (see Figs. 1 and 2). Naturally the hot boundary should normally be selec-

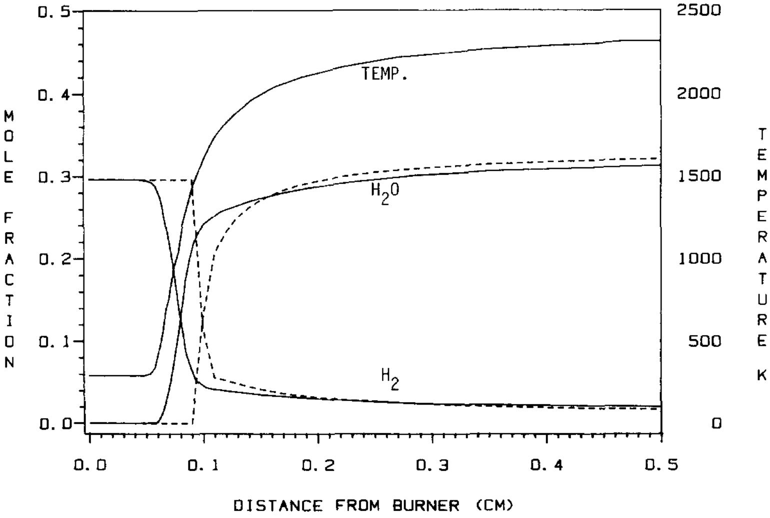

FIG. 1. Starting profiles (broken lines, this study) for H_2 and H_2O calculated with a kinetics approximation of a steady state solution. Temperature and mass flow for the kinetic approximation were taken from the steady state solution with diffusion included. The system is H_2/air at 1 atm. The solid lines represent a solution of the system with diffusion included [34].

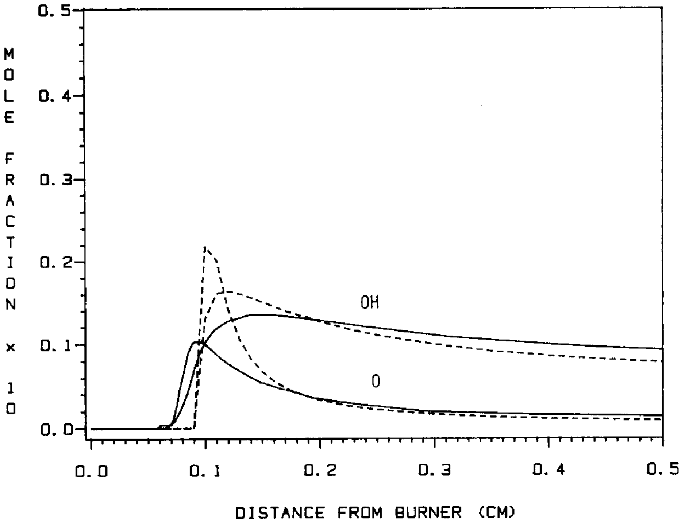

FIG. 2. Starting profiles (broken lines, this study) for O and OH calculated with a kinetics approximation of a steady state solution. Temperature and mass flow for the kinetic approximation were taken from the steady state solution with diffusion included. The system is H_2/air at 1 atm. The solid lines represent a solution of the system with diffusion included [34].

ted so that the species show very low space gradients at that point. The values calculated at the hot boundary, which are approximately the equilibrium values, are used as fixed boundary values together with the cold boundary values already discussed during the time-dependent calculations which are discussed below.

3.2. *Solution of the Time-Dependent Equation System*

For the time-dependent approach to the steady state solution at each time step and each grid point we solve a system of ordinary differential equations. In the time step the first order implicit method is applied for maximum stability

$$X_i(t+\Delta t) - X_i(t) = X_i'(t+\Delta t) * \Delta t \tag{16}$$

with the derivative expanded as

$$X_i'(t+\Delta t) = X_i'(t) + \sum_j \frac{\partial X_i'}{\partial X_j} * (X_j(t+\Delta t) - X_j(t)). \tag{17}$$

The final linear equation system is then

$$\sum_j \left(\delta_{ij} - \frac{\partial X_i'}{\partial X_j} * \Delta t \right) (X_j(t+\Delta t) - X_j(t)) = X_i' * \Delta t. \tag{18}$$

For the transport term, an explicit evaluation is made and the partial derivatives are therefore only evaluated for the kinetics contribution. The linear equation system is solved by the least square subroutine LLSQF from IMSL Inc., Houston, Texas.

With the rather simple expression for the kinetics contribution the partial derivatives can easily be derived analytically. In a large reaction mechanism containing many species, as in the Westbrook–Dryer model for methanol/air combustion [12] which involves 26 species in 84 reaction pairs, a large proportion of the computer time is spent evaluating these partial derivatives by conventional numerical differentiation. With the analytical derivation we found that the computer time was reduced by a factor of 2.

3.3. *Time Step and Convergence Measure*

The time-dependent method simulates a physical relaxation of a reactive diffusive system towards steady state. The time step for an explicit diffusion model is restricted by the grid point distances selected and the diffusion coefficients of the different species according to theory [14, 23, 33]. This theory predicts that the time step $dt = (dx)^2/2D$, where dx is the minimum grid distance and D is an averaged diffusion coefficient. We used this formula for estimating the maximum time step.

The relative error, obtained by comparing a centered solution and an implicit solution, also gives information on how to decide on the practical time step; a significant increase in the relative error signals a time step which is too large. We also use the time gradient of the species

$$\sum_i |X_i' * M_i| \tag{19}$$

averaged over all grid points to indicate if the time step is small enough. A time step which is too large means that the calculated profiles move away from the steady solution. This is indicated by a corresponding increasing time gradient. The time gradient expresses the change in mass per time unit of the combustion system at a certain time, and should decrease during the relaxation to the steady state. Consequently, we also use the time gradient as a convergence measure.

We expect that the elapsed total physical time, which is the sum of all time steps taken, at convergence (steady state) should be comparable to the flow-time along the grid.

3.4. *Grid System with Successive Grid Refinements*

We use a nonuniform grid without any form of transformation of the space coordinate. The grid points are put in manually and concentrated in the flame zone to increase the resolution of wings and peaks of the species. If one wants to automate this procedure a simple way is to use the temperature profile for grid point selection [19]. It appears that the more accurate method implemented by Smooke [7] could also be used. By starting with a coarse grid we can reach the steady state with large time steps (see the formula for the maximum time step) at a low computational cost.

To reach a high resolution of the concentration profiles successive grid refinements are used [7]. The first coarse grid is used as an approximation for a second grid. The mole fractions of the species at new grid points are evaluated by a linear interpolation of the molfractions at the previous grid points. This induces a higher time gradient initially, which within 15–25 time steps relaxes to nearly the steady state value on the previous grid. The calculations are continued until the time gradient is below a certain value which corresponds to the target convergence level for that grid. The necessary physical relaxation time for the calculation on the second grid is only a fraction of the relaxation time on the coarse grid, if new points are inserted only in the flame zone. This procedure of grid refinements is repeated until the species profiles are visually independent of the change in grid refinement.

4. Computational Results from Test Flame Calculations

The test flames were selected to make it possible to compare our results with similar computations obtained by some well-known and frequently used computer programs [7, 19]. Many other excellent studies have been made concerning modeling of laminar flames (see, for example, [13–18, 19–24]). Here the main comparison is with the program documented by Smooke. It is a steady state program and Smooke estimated [7] that this program (method) is 5–10 times faster than traditional time-dependent computer programs. We made a computational study on the H_2/air and CH_4/O_2 systems studied by Smooke. Previously we made a comparison [5] with the time-dependent program by Lund [19] and used extensively by Westbrook [31]. In that study of CH_3OH/air we also compared our calculations with experimental results.

4.1. *H_2/Air Calculations*

Smooke, Kee, and Miller have published detailed calculated concentration profiles for H_2/air at 1 atm and stoichiometric conditions giving a mass flow of 0.1550 g/cm^2, s [34]. In the mechanism they used 8 species H_2, O_2, N_2, H, O, OH, H_2O, and HO_2 involved in 11 reaction pairs [17].

We modeled the H_2/air system above using the same grid distances, reaction mechanism, and binary diffusion coefficients as Smooke *et al.* [34]. As we do not calculate mass flow and temperature in our program it was necessary to use the corresponding values from Smooke *et al.* [34]. Unlike Smooke *et al.* [34], we used no thermal diffusion and no multicomponent molecular diffusion model in our calculations. However, we found that thermal diffusion (using the model recommended by Coffee and Heimerl [30]) did not affected the results in a preliminary calculation. (In Section 5, the effect of diffusion models on accuracy is discussed.) The flow-time calculated in the kinetics approximation was 0.9 ms for the 1-cm-long grid used.

A constant time step according to the formula in Section 3.3 could be used except for the first 10 time steps on the first grid, where a factor of 3 lower time step has to be used. The relaxation time to reach steady state was about 0.6 ms. On the first grid we also did a calculation with a constant starting approximation for H_2, O_2, and N_2 together with an assumed temperature profile and mass flow. For that case the relaxation time was about 40% longer than the corresponding calculation started with the kinetics approximation.

On the final grid 54 points were used with 28 points in the flame zone giving a minimum interval of 0.0025 cm and a maximum time step about 0.1 μs. The flame zone is defined [7] as the z-interval where temperature increases from 10 to 90% of the total temperature increase. In our calculations the execution time on an IBM 3033N was 367 CPU s after 1200 time steps and at convergence the time gradient was 4.8 g/s on the final grid used. We reached a true convergence at that gradient as confirmed by a subsequent calculation for a physical time of 0.2 ms with no changes in the profiles. With the final grid used all the way from the kinetics approximation we needed 6000 time steps to reach the same convergence level corresponding to a time gradient of 4.9 g/s. The total execution time for this conventional method without grid refinements was 2058 CPU s. Figures 3 to 6 give our results and the corresponding results from Smooke *et al.* [34]. The agreement is excellent and the remaining minor differences are within the uncertainties expec- [illegible]

Smooke *et al.* gave an execution time of [illegible] CPU s on a CRAY 1S computer for 53 points with 22 points in the flame zone [34]. In that calculation temperature and mass flow were also calculated. Recently Smooke reran the calculation with

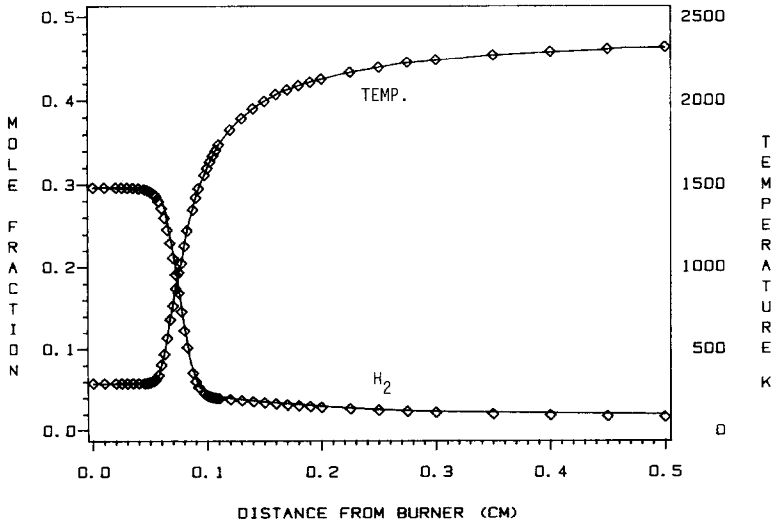

FIG. 3. Calculated profiles of H_2 for an adiabatic hydrogen-air flame at 1 atm. The solid line is H_2 calculated by Smooke *et al.* and the data points represent our corresponding profile. The temperature profile calculated by Smooke (solid line) and our assumed profile (symbol) taken from Smooke *et al.* is also shown.

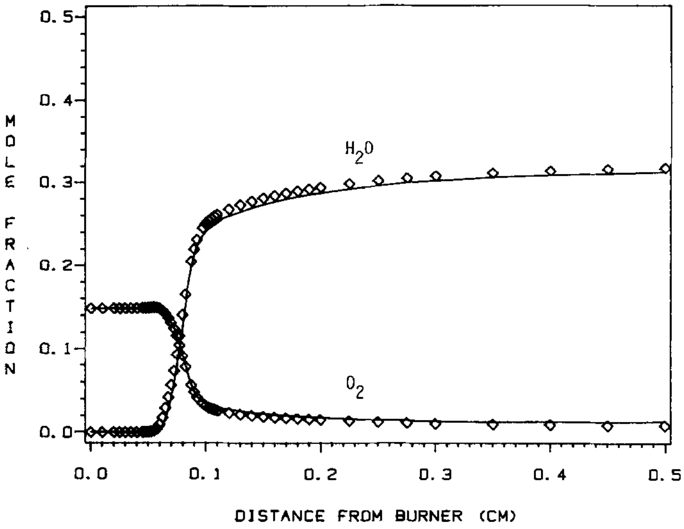

FIG. 4. Profiles of O_2 and H_2O, calculated by us (symbols) and by Smooke *et al.* (solid lines) for an adiabatic hydrogen-air flame at 1 atm.

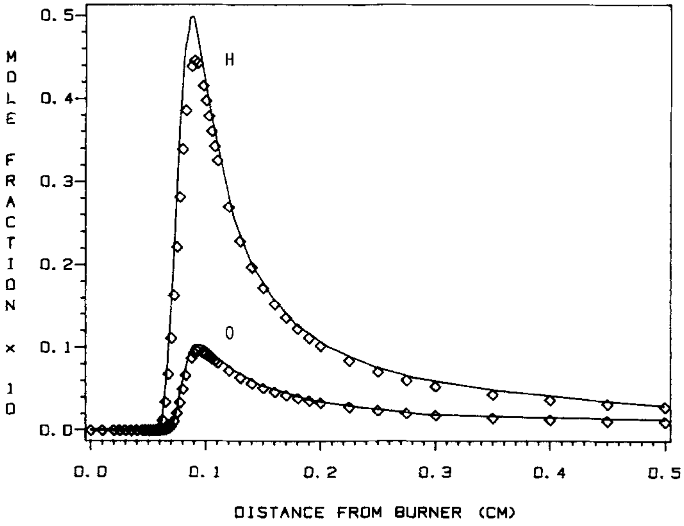

FIG. 5. Profiles of O and H, calculated by us (symbols) and by Smooke *et al.* (solid lines) for an adiabatic hydrogen-air flame at 1 atm.

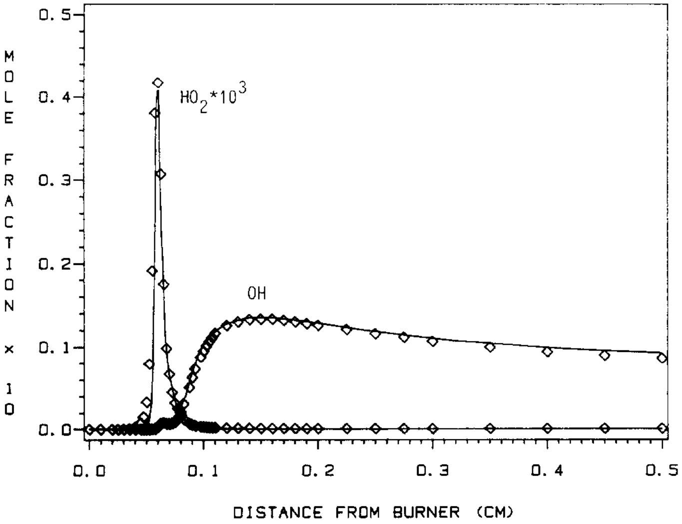

FIG. 6. Profiles of OH and HO_2, calculated by us (symbols) and by Smooke *et al.* (solid lines) for an adiabatic hydrogen–air flame at 1 atm.

temperature and mass flow specified using the same diffusion model we used [36]. The resulting execution time was 13 CPU s on the CRAY computer. For our time-dependent flame program we estimate that an IBM 3033N computer is at least a factor 5 slower than a CRAY 1S.

4.2. *CH_4–O_2 Calculations*

Peeters and Mahnen studied experimentally CH_4/O_2 flames [37]. The initial gas mixture consisted of 9.5 mole percent CH_4 and 90.5 mol% O_2 with a flame velocity of 67 cm/s, at a cold gas temperature 293 K, and a pressure of 0.052 atm giving a mass flow of 0.00448 g/cm^2, s. Smooke modeled that flame on a 5-cm-long grid using the experimental conditions above and the experimental temperature profile together with the mechanism compiled by Tsatsaronis [17]. The mechanism consists of 13 species, the H_2/O_2 species H_2, O_2, H_2O, H, O, OH, and HO_2 and the hydrocarbon species CO, CO_2, CHO, CH_2O, CH_4, CH_3. The species are involved in 29 pairs of forward/backward elementary reactions.

We modeled the CH_4/O_2 system above using the same mass flow, temperature profile, and reaction mechanism as Smooke. A 10-cm-long grid was used imposing vanishing gradients in the hot boundary region. Diffusion was allowed into the burner by extrapolating the temperature profile 0.2 cm into the porous burner. This was done to mimic the cold boundary conditions used by Smooke. The mechanism of Tsatsaronis is rather artificial and provides no ignition reactions, therefore it was complemented with reaction number 22 $CH_4 + O_2$, from the Westbrook mechanism [31], during the kinetics approximation calculation [2].

The flow time on the 10-cm-long grid was 25 ms and the relaxation time towards steady state was about 25 ms. Five successive grids were used resulting in an execution time of 490 CPU s on an IBM 3033N and with a total of 1000 time steps. On the last grid the convergence time gradient was 0.39 g/s. On the final grid 51 points were used with 23 of them in the flame zone and a minimum grid interval of 0.0125 cm and a maximum time step of about 0.15 μs. For that flame our results are shown in Figs. 7–9 together with the corresponding results from Smooke. In that study a multicomponent diffusion model without thermal diffusion was used. Smooke used 45 points with 24 of them in the flame zone and the calculation took 41 CPU s on a CRAY 1S computer. For a trace diffusion model similar to our diffusion model his execution time was 24 CPU s.

4.3. *CH_3OH–Air Calculations*

We modeled stoichiometric methanol/air flames in a separate study [5] so we just summarize the results. Westbrook and Dryer modeled CH_3OH/air flames with 26 species and 84 pairs of elementary reactions [31]. We reproduced their results at 0.1 atm within a few percent when we used their empirical diffusion coefficients. When using a more fundamental diffusion coefficient description, as defined in Eq. (12) their profiles were much broader. This is due to the fact that the Westbrook–Dryer diffusion coefficients were a factor of 3–4 too high. The convergence time gradient was about 1 g/s at steady state.

In this study we made experiments on methanol/air combustion at 0.133 atm. For that study we used the conventional diffusion coefficients complemented with a

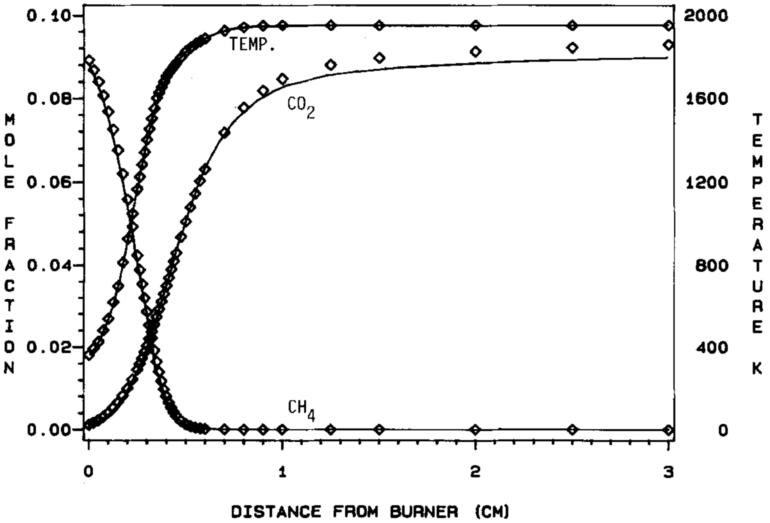

FIG. 7. Profiles of CH_4 and CO_2, calculated by us (symbols) and by Smooke *et al.* (solid lines) for a methane–oxygen flame at 0.052 atm studied by Peeters and Mahnen [19]. In the calculations the same smoothed extrapolation of the temperature profile was used, calculated by us and by Smooke *et al.*

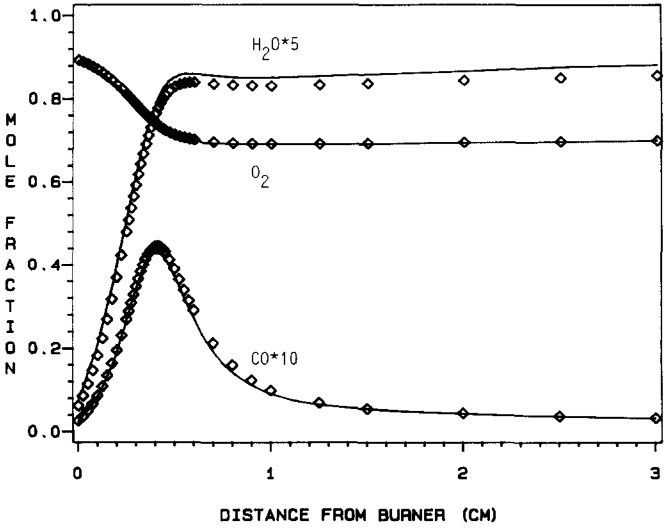

FIG. 8. Profiles of O_2, H_2O and CO, calculated by us (symbols) and by Smooke *et al.* (solid lines) for a methane–oxygen flame at 0.052 atm.

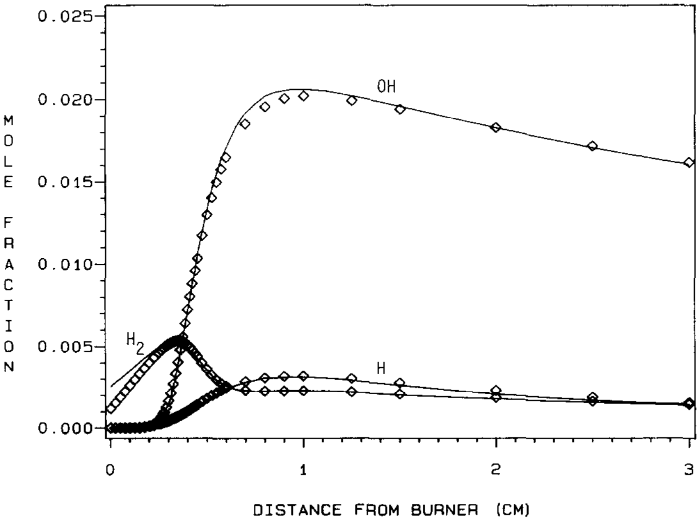

FIG. 9. Profiles of OH, H_2, and H, calculated by us (symbols) and by Smooke *et al.* (solid lines) for a methane–oxygen flame at 0.052 atm.

simplified Westbrook–Dryer mechanism. In general the agreement was good. The experimental results showed steeper gradients compared to the computations. This could by explained by nonlinear cooling of the flame by the sampling cone.

5. DISCUSSION

5.1. *Physical–Chemical Simplifications*

We used some physical simplifications in the program: diffusion was modeled as trace diffusion, the diffusion coefficients were calculated in a simple way, thermal diffusion was not included, and the diffusion velocities were not corrected for mass conservation [7, 2]. All of these simplifications are small compared to uncertainties in experimental measurements. Furthermore we reproduced results of Smooke [7] and of Smooke *et al.* [34] with minor discrepancies. Their transport model is more sophisticated, including multicomponent diffusion, more detailed description of diffusion coefficients, thermal diffusion (for the H_2/air test flame), and a constant average diffusion velocity correction. We checked numerically the effects of these details and found that they have no visible effects for the flames we have studied. However, we want to point out that for many flame systems more complex transport models may be needed, for example, if no component is in excess.

The effect of thermal diffusion was insignificant in our study of H_2/air. This was to be expected from the Warnatz study [22, 23] of H_2/air combustion indicating the necessity for thermal diffusion mainly in the case of rich flames. He found that inclusion of thermal diffusion slightly lowered the flame velocity ($\sim 5\%$ for a flame with 45% H_2 in the unburnt gas). It is to be emphasized that we made our study on the assumptions of known flame velocity and temperature and on a stoichiometric flame. Through known temperature and mass flow the first order effects of the transport model are taken into account.

In conclusion, the simple diffusion model we used here is accurate for the test flames above with known temperature and mass flow. We believe that this diffusion model is rather accurate for similar systems, for example, the important class of hydrocarbon–air flames.

5.2. *Starting Profiles*

The starting approximations can influence the execution time in at least three different ways. First, if the physical ignition time is long, say many milleseconds, this would create an unacceptable long execution time with a small time step of 0.1 μs. This is a problem if one starts with constant fuel and oxidant in combination with a low initial energy. Second, even after ignition is secured, the maximum time step allowed can be very small and hard to predict [19, 16]. It appears that the reduced maximum time step is caused by increased importance of the chemical kinetics. Such a change would reduce the time step according to the stability criterion. This problem has sometimes been solved by providing initial broad species profiles [22].

Third, a good starting approximation can reduce the relaxation time to steady state by 35% as we found in this study.

The procedures found in the literature for good starting profiles necessitate estimation of the widths and maxima of the profiles. By using these parameters in certain functions *S*-shaped and Gaussian initial profiles are formed. By using an initial chemical kinetics calculation to provide starting profiles and hot boundary conditions we could minimize work on estimations. This initial solution is instructive and has a direct physical meaning. Normally stable numerical conditions are reached after less than 100 time steps. The flame front instability is smoothed, and we can take fixed large time steps for the rest of the calculations. A modest investment in execution time with a rough grid can afterwards provide a good approximation of the steady state.

5.3. *Hot Boundary Conditions*

We calculate the hot boundary values by doing a chemical kinetics calculation with the experimental temperature profile. The hot boundary values are kept fixed during the computations. For an adiabatic flame experiment our approach converges towards the procedure of Smooke *et al.* [34, 35], who use zero gradient values for adiabatic flames at the hot boundary. For both methods it is important to use a grid long enough that the species gradients are small at the boundary.

For the calculation on the experimental flame system CH_4/O_2 above with fixed hot boundary values a 10-cm-long grid was used. At the distance 5 cm used by Smooke the important species OH still has a significant gradient. However, Smooke compensated for the shorter grid distance by allowing the concentrations at the last calculated grid point to vary in the approach towards steady state [24]. We used that approach initially, but we had stability problems especially for narrow computing intervals. By using fixed hot boundary values numerical stability was increased.

5.4. *Time Step and Convergence*

We found that a convenient method was to use a fixed time step. As discussed above the theoretical formula for determining the maximum time step $dt = (dx)^2/2D$ was approximate within a factor of 2 for the H_2/air system at a pressure of 1 atm, except for the initial 10 time steps on the first grid. Possibly the discrepancies above are due to a contribution from a chemical kinetics term not used in the derivation of the formula [16, 33]. For the CH_4/O_2 system at a pressure of 0.05 atm we use a constant time step all the time. In both cases D is about $25/p$ cm^2/s with p the pressure in atmospheres.

Time-dependent calculations simulate a relaxation towards the stationary state. We found that this relaxation time is about equal to the flow time. We found that the H_2/air, CH_3OH/air, and CH_4/O_2 converged at time gradients of about 5, 1, and 0.4 g/s, respectively. It appears that this is related to the difference in pressure 1, 0.1, and 0.05 atm, respectively.

5.5. *Successive Grid Refinements*

The relaxation time divided by the maximum time step gives the minimum number of necessary time steps. The execution time is directly proportional to the number of grid points multiplied by the number of time steps. Consequently, to reduce the execution time we calculate to a higher degree of convergence (lower time gradient) on the initial grid than we want on the refined grid, since the calculation cost is lower for the coarse grid with fewer grid points. Also, longer time steps are allowed on the coarser grid due to the longer distances used between the grid points.

If the conventional time-dependent method is used, with a nonuniform density of grid points a small time step 1 or 0.1 μs must be used all the time, and it can be necessary to take 5000 or more time steps to reach the true steady state on large dense grids as in our examples, giving a long relaxation time and a short maximum time step. In the H_2/air study we used only a total of 1200 time steps thanks to the use of successive grid refinement. With a maximum time step of 0.1 μs on the last grid 6000 time steps were necessary to reach the steady state without grid refinements as the relaxation time was 0.6 *m*s. We estimate that more than 100,000 time steps would be necessary to reach steady state for the CH_4/O_2 test flame with the same grid if we used the conventional time-dependent method. We estimated that figure because a calculation using the fine grid from the start was too expensive due to the long execution time necessary (more than 24 h on an IBM 3033). In contrast, we needed only 1000 time steps using successive grid refinements. In addition the mean cost per time step is lower for a method based on successive grid refinements compared to the conventional method, in which the number of grid points is constant and at maximum throughout all of the calculations.

5.6. *Execution Times of Our Time-Dependent Version*

Smooke stated that steady state methods have the potential of solving the flame equations 5–10 times faster than time-dependent methods. First, he reasoned that the number of iteration steps necessary in the steady state method are less than in the time-dependent method. Second, Smooke assumed that the work for a steady state Newton iteration is about the same as for a time step. Our results confirm Smooke's opinion that the steady state method is at least 5 times faster than the time-dependent method used in a conventional way. Our time-dependent calculations with successive grid updating show execution times longer by a factor of two than the original steady state calculations done by Smooke *et al.* in 1981 [34, 35] and by Smooke in 1982 [7]. The number of time steps in our calculations is about 20 times higher than the number of iteration steps in the original steady state calculations by Smooke. This means that the cost per step is about 10 times higher in the steady state calculation than in our time-dependent calculation.

Here we want to mention a few factors, which can partly explain our low cost per time step. Our simple number representation of the reactants in contrast to the commonly used matrix representation in combination with analytical calculation of the partial derivatives reduces the execution time per timestep by a total factor of 3–4. The diffusion model is simple and fast. Smooke reran his calculations with the same diffusion simplifications as we made [36], with the result that the diffusion model version used here was a factor 2–3 faster than the more general diffusion model documented previously by Smooke [7]. Clearly, the steady state method implemented by Smooke [7] with a simple diffusion model as in this study is a factor 4 faster than our present implementation of the time-dependent method. However, we believe that our time-dependent version can be further optimized. The reason is that the most of the execution time comes from calculations on the last grids showing very small physical changes per time step. The corresponding time derivatives are changing only slowly per time step offering opportunities in computational saving.

In conclusion the present implementation of the steady state method is faster than our time-dependent version. However, the use of successive grid refinements in time-dependent calculations decreased the difference in execution times. In addition, the time-dependent flame programs are robust [7]. Also time step restriction and convergence measure can be intuitively understood and related to minimum grid interval and the flow time along the grid, respectively.

6. Conclusions

Successive grid refinements with a convergence reached on each grid can reduce the total execution time of time-dependent methods by about a factor of 10 or more. However, our time-dependent method is still a factor of 4 times slower than the steady state method with a similar diffusion model. These two seemingly very different numerical method give the same concentration profiles to within 1%.

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