Numerical investigation of thermal diffusion influence on soot formation in ethylene/air diffusion flames

Guo, H.; Liu, F.; Smallwood, G.J.; Gulder, O.L.

Publisher's version / Version de l'éditeur:

The Tenth Annual Conference of the CFD Society of Canada: CFD 2002: proceedings, June 9-11, Windsor, Ontario, pp. 180-185, 2002

MARIA CLANCY
DGO
INST FOR CHEM PROCESS & ENVIR TECH
NATIONAL RESEARCH COUNCIL CANADA
M-12, ROOM 141, 1200 MONTREAL RD.
OTTAWA, ON K1A 0R6
CANADA

ORDER NUMBER:	**CT-07782490-9**
Account Number:	WR001844
Delivery Mode:	XLB
Delivery Address:	
Submitted:	2009/03/04 09:34:13
Received:	2009/03/04 09:34:13
Printed:	2009/03/09 10:56:50

Extended	**Conference**	**Internet - transcribed**	**CANADA**

Client Number:	MARIA CLANCY MARCH 2, 2009 # 60
Title:	**PROCEEDINGS OF THE 10TH ANNUAL CONFERENCE OF THE CFD SOCIETY OF CANADA, WINDSOR, ON**
Author:	GUO, H.; LIU, F.; SMALLWOOD, G.J.; ET AL
Vol./Issue:	10TH, JUN
Date:	2002
Article Title:	NUMERICAL INVESTIGATION OF THERMAL DIFFUSION INFLUENCE ON SOOT FORMATION IN ETHYLENE/AIR FLAMES

Estimated cost for this 6 page document: $0 document supply fee + $0 copyright = $0

NUMERICAL INVESTIGATION OF THERMAL DIFFUSION INFLUENCE ON SOOT FORMATION IN ETHYLENE/AIR DIFFUSION FLAMES

Hongsheng Guo, Fengshan Liu, Gregory J. Smallwood
Combustion Research Group
National Research Council of Canada
1200 Montreal Road, Ottawa, Ontario K1A 0R6
Email: hongsheng.guo@nrc.ca

Ömer L. Gülder
Institute for Aerospace Studies, University of Toronto
4925 Dufferin Street, Toronto, Ontario M3H 5T6

ABSTRACT

A numerical investigation of the thermal diffusion influence on soot formation in coflow ethylene/air laminar diffusion flames has been conducted. Detailed reaction mechanism and complex thermal and transport properties were employed. The fully elliptic governing equations were solved. Radiation heat transfer from the flames was obtained by the discrete ordinate method coupled to a SNBCK-based wide band model. A simplified two-equation soot model was used. The thermal diffusion velocities were calculated according to the thermal diffusion coefficients that were evaluated based on multicomponent properties.

The results show that thermal diffusion does affect soot formation in ethylene/air diffusion flames. Soot volume fraction is overpredicted if thermal diffusion is neglected. The influence of thermal diffusion is enhanced if there are lighter species, such as helium, in the fuel or the air streams. For instance, the peak integrated soot volume fraction doubles if thermal diffusion is not taken into account in the simulation for the flame with helium addition to the air stream.

INTRODUCTION

Thermal diffusion (Soret effect), caused by temperature gradients in a mixture, gives an additional term in the diffusion velocity of a chemical species. It tends to draw lighter molecules to hot regions and to drive heavier molecules to cold regions of the mixture.

In a pioneering work, Dixon-Lewis [1] observed that the thermal diffusion flux for hydrogen could be of the same order of magnitude as the ordinary diffusion flux in a hydrogen/air flame. The same observation was made by Greenberg [2] in the study of a one-dimensional hydrogen/air flames using a one-step reaction model and phenomenological expressions for the thermal diffusion coefficients. Later it was found by Warnatz [3] that the laminar flame speeds of both lean and rich hydrogen/air flames were lower when thermal diffusion was considered, although only the thermal diffusion fluxes of atomic and molecular hydrogen were considered in the simulation. In the study of vortex-flame interactions in hydrogen jet diffusion flames, Hancock et al. [4] showed that thermal diffusion effect couldn't be neglected in the numerical simulation. A recent study by Ern et al. [5] showed that thermal diffusion is important not only for the prediction of structures of hydrogen/air and methane/air Bunsen flames, but also for the prediction of NO in a counterflow methane/air flame. Being different from the result of [3], the study of [6] indicated that for hydrogen/air flames, while the speeds of lean flames were lower, those of rich flames were higher when thermal diffusion was considered. The influence of thermal diffusion on the flame speeds of methane/air flame is negligible. More recently, Williams [7] revealed that thermal diffusion effect resulted in an increase in the predicted extinction strain rate of methane/air counterflow flame.

In spite of the importance of thermal diffusion, little attention has been paid to the influence of thermal diffusion on soot formation process. It was totally neglected in some studies [8-10], while only the thermal diffusion of light species (such as H_2 and H) was taken into account in some other studies [11]. To our knowledge, the relative influence of thermal diffusion on soot formation has not been reported previously in the literature.

In the present paper, soot formation processes in a pure ethylene/air and four helium or argon diluted ethylene/air coflow laminar diffusion flames were simulated. The objective is to investigate the relative influence of thermal diffusion on soot formation. We employed the primitive variable method in which the fully elliptic governing equations were solved with detailed gas-phase chemistry and complex thermal and transport properties. The effects of soot inception, growth and oxidation on gas-phase chemistry were considered. For the soot kinetics process, a simplified two-equation soot model was used.

NUMERICAL METHODS

The flames studied are coflow axisymmetric laminar diffusion flames. They are generated with a burner [12] in which the fuel stream flows from a 10.9mm inner diameter vertical tube, and the oxidant stream flows from the annular region between the fuel tube and a 100mm diameter concentric tube. The wall thickness of the fuel tube is 0.95 mm.

Gas Phase Governing Equations

The governing equations of gas phase have been described elsewhere [13]. The radiation heat transfer was calculated by the discrete-ordinates method coupled to a SNBCK based wide band model [14]. For the sake of brevity, only the transport equations of gas species are given here. They are

$$\rho v\frac{\partial Y_k}{\partial r}+\rho u\frac{\partial Y_k}{\partial z}=-\frac{1}{r}\frac{\partial}{\partial r}\left(r\rho Y_k V_{kr}\right)-\frac{\partial}{\partial z}\left(\rho Y_k V_{kz}\right)+W_k\omega_k$$

$$k=1,\ 2,\ \ldots,\ KK \qquad (1)$$

where u and v are the velocities in axial (z) and radial (r) directions, respectively; ρ the density of the mixture; W_k the molecular weight of the kth gas species; ω_k the mole production rate of the kth gas species, and Y_k the mass fraction of the kth gas species; Quantities V_{kr} and V_{kz} are the diffusion velocities of the kth gas species in r and z directions; and KK the total gas phase species number.

The diffusion velocity consists of three terms: ordinary diffusion, thermal diffusion and correction diffusion velocities. Therefore:

$$V_{kx_i}=V_{okx_i}+V_{Tkx_i}+V_{cx_i}, \qquad k=1,2,\ldots,KK, \qquad x_i=r,\ z \qquad (2)$$

Since the current study concentrates on the relative influence of thermal diffusion, the ordinary diffusion velocity V_{okx_i}, caused by concentration gradient, was obtained by the approximate mixture-average formulation, i.e.

$$V_{okx_i}=-\frac{1}{Y_k}D_k\frac{\partial Y_k}{\partial x_i} \qquad k=1,2,\ldots,KK, \qquad x_i=r,\ z \qquad (3)$$

Quantity V_{Tkx_i} is the thermal diffusion velocity, whose influence will be investigated in the present paper, in x_i direction for the kth gas species. It was obtained by [15]:

$$V_{Tkx_i}=-\frac{D_k^T}{\rho Y_k}\frac{1}{T}\frac{\partial T}{\partial x_i} \qquad k=1,2,\ldots,KK, \qquad x_i=r,\ z \qquad (4).$$

where D_k^T is the thermal diffusion coefficient obtained by the method given in [15].

The correction diffusion velocity V_{cx_i} is used to ensure that the net diffusive flux of all gas species and soot is zero [15].

Quantity D_k in Eq. 3 is related to the binary diffusion coefficients through the relation

$$D_k=\frac{1-X_k}{\sum_{j\neq k}^{KK} X_j/D_{jk}}, \qquad k=1,2,\ldots,KK \qquad (5)$$

where X_k is the mole fraction of the kth species, and D_{jk} is the binary diffusion coefficient.

Soot Model

Two transport equations were solved for soot mass fraction and number density. The acetylene (C_2H_2) based semi-empirical soot inception, surface growth and oxidation models [16, 17] were used to describe the soot process in the flame.

The details of the calculations of soot inception, surface growth and oxidation can be found from our previous work [13] and the references cited there.

Numerical Model

The governing equations were discretized using the control volume method. The SIMPLE numerical scheme [18] was used to deal with the pressure and velocity coupling. The diffusion and convective terms in the conservation equations were respectively discretized by the central and upwind difference methods. The discretized equations of gas species, soot mass fraction and soot number density were solved in a fully coupled fashion on every grid to speed up the convergence process [19], while those of momentum, energy and pressure correction were solved using the tri-diagonal matrix algorithm.

The computational domain covers an area from 0 to 3.0 cm in the radial direction and 0 to 11.0 cm in the axial direction. The inflow boundary corresponds to the region immediately above the fuel nozzle.

The chemical reaction mechanism used is essentially from GRI-Mech 3.0 [20], with the

removal of all the reactions and species related to NO_X formation. All the thermal and transport properties were obtained by using the database of GRI-Mech 3.0 and the algorithms given in [15, 21].

RESULTS AND DISCUSSIONS

Five flames were studied. They are: pure ethylene/air flame, and the flames with 30% helium addition to fuel, 30% helium addition to air, 30% argon addition to fuel and 30% argon addition to air. The volume flow rates of fuel and air were kept constant for all the flames, while helium or argon was added to fuel or air in the second to fifth flames.

For every flame, two simulations, one with thermal diffusion and the other without thermal diffusion, were conducted.

Pure Ethylene/Air Flame

We first investigate the influence of thermal diffusion on the prediction of soot formation in pure ethylene/air flame by comparing the results of simulations with and without thermal diffusion. Fig. 1 depicts the integrated soot volume fractions for pure ethylene/air flame (in all the figures, TD represents thermal diffusion). It is noted that soot volume fraction was overpredicted when thermal diffusion was neglected. The peak integrated soot volume fraction is about 2.6% higher if thermal diffusion is neglected.

Figure 2 shows the thermal diffusion factors, defined as the ratio of thermal diffusion coefficient (D_k^T) to the product of mixture density and species mass fraction (ρY_k), of several main species and radicals at Z (axial distance) = 1.0 cm. The radial temperature profile at Z = 1.0 cm is also shown for completeness. It indicates that the thermal diffusion factors of species H_2O, H_2, H, O and OH are negative, while those of CO_2 are positive. Therefore thermal diffusion tends to draw the lighter species, such as H_2O, H_2 and H, to the hotter region and to drive the heavier species, such as CO_2, to the colder region. Examples are given in Fig. 3 for H_2 and CO_2.

The variations of species concentrations introduce the disparities in the flame temperatures, and thus finally cause the discrepancies of soot inception and surface growth processes, as shown in Fig. 4. Accordingly soot volume fractions are higher in most of radial region for the simulation without thermal diffusion.

The situations at other axial heights are similar to that at Z = 1.0 cm. Therefore the integrated soot volume fractions are overpredicted if thermal diffusion is not taken into account.

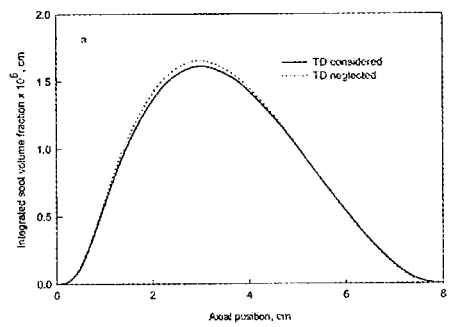

Fig. 1 Integrated soot volume fractions of pure ethylene/air flame.

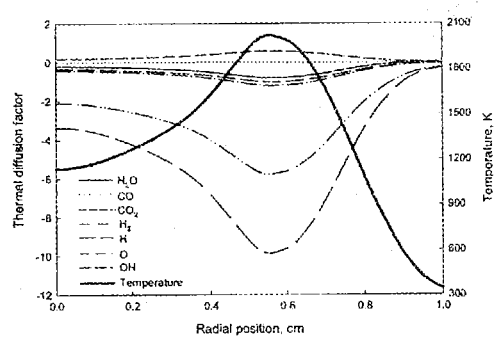

Fig. 2 Thermal diffusion factors at Z = 1.0 cm.

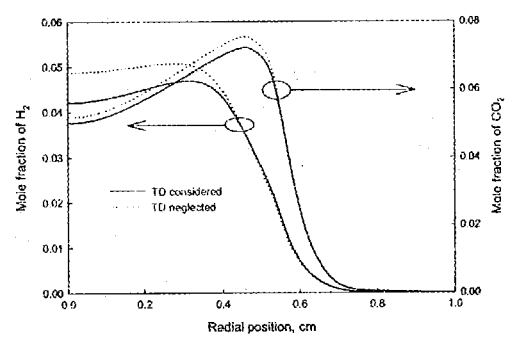

Fig. 3 Radial profiles of H_2 and CO_2 at Z = 1.0 cm.

Flame with 30% Helium Addition to Fuel

Figure 5 compares the integrated soot volume fractions of the simulations with and without thermal diffusion when 30% helium is added to fuel. Similar to pure ethylene/air flame, soot volume fraction is overpredicted if thermal diffusion is neglected. However, the influence is increased compared to pure ethylene/air flame. The peak integrated soot volume fraction is overpredicted by 5.3% for this flame.

In addition to those variations caused by thermal diffusion for pure ethylene/air flame, helium

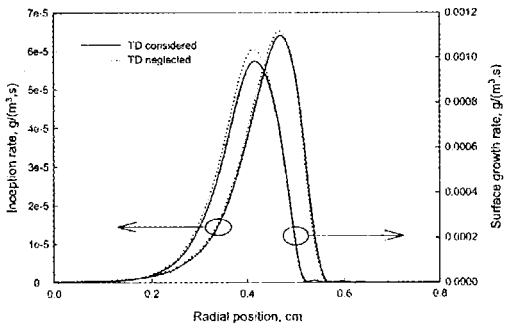

Fig. 4 Inception and surface growth rates of pure ethylene/air flame at Z = 1.0 cm.

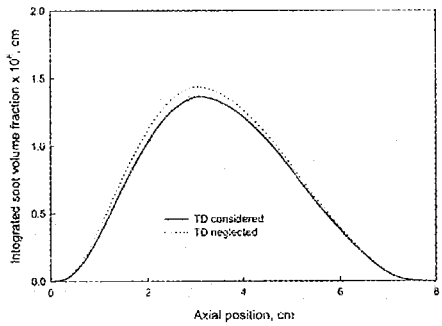

Fig. 5 Integrated soot volume fractions of the flame with 30% helium addition to fuel.

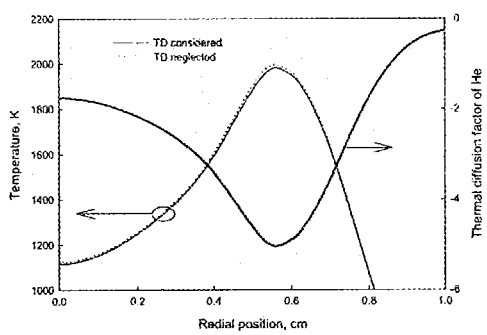

Fig. 6 Profiles of temperature and thermal diffusion factors of helium at Z = 1.0 cm.

is driven to the reaction (hot) zone due to its negative thermal diffusion factors, as shown in Fig. 6, in this flame. Therefore the variations of temperatures in the reaction zone caused by thermal diffusion are further increased, due to the addition of helium to fuel. Accordingly the influence of thermal diffusion on soot formation is greater in this flame than in pure ethylene/air flame.

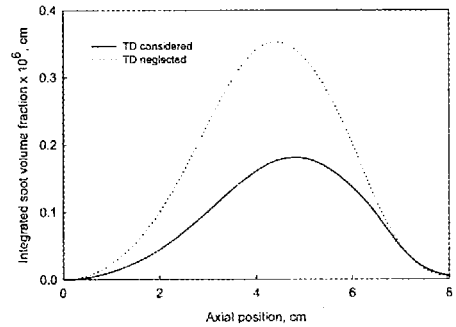

Fig. 7 Integrated soot volume fractions of the flame with 30% helium addition to air.

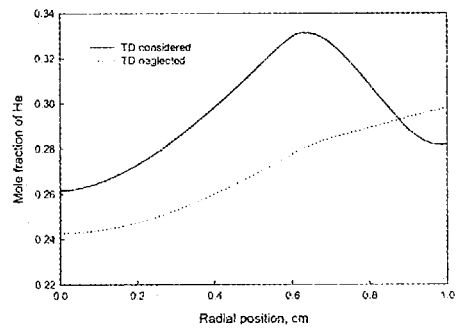

Fig. 8 Mole fraction of helium at Z = 1.0 cm of the flame with 30% helium addition to air.

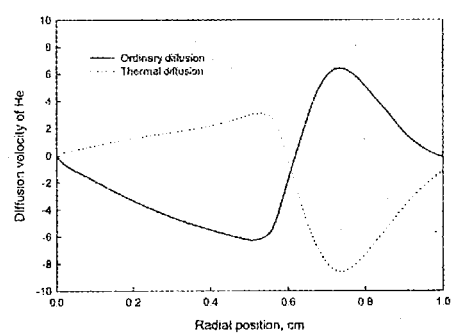

Fig. 9 The diffusion velocities of helium at Z = 1.0 cm of the flame with 30% helium addition to air.

Flame with 30% Helium Addition to air

Figure 7 shows the influence of thermal diffusion on integrated soot volume fractions of the flame with 30% helium addition to air. Thermal diffusion effect becomes much more significant for this flame. The peak integrated soot volume fraction almost doubles if thermal diffusion is neglected in the simulation. Again this is due to the moving of helium to the hot region because of thermal diffusion.

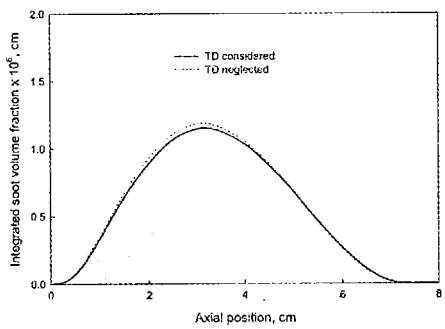

Fig. 10 Integrated soot volume fractions of the flame with 30% argon addition to fuel.

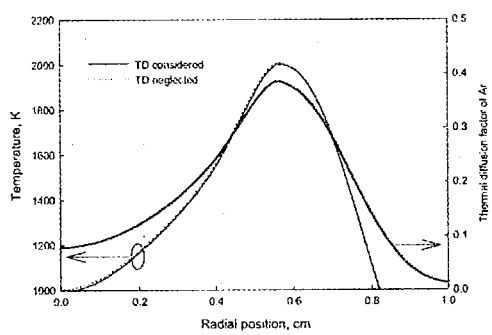

Fig. 11 Profiles of temperature and thermal diffusion factors of argon at Z = 1.0 cm for the flame with 30% argon addition to fuel.

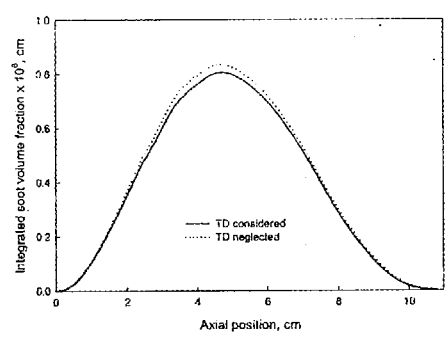

Fig. 12 Integrated soot volume fractions of the flame with 30% argon addition to air.

Figure 8 shows that the concentrations of helium in the reaction (hot) zone for the simulation with thermal diffusion are much higher than those of the simulation without thermal diffusion are. A more interesting phenomenon is that the concentrations of helium in reaction zone are higher than those in the periphery region. Since helium is added to the air, this phenomenon implies that the thermal diffusion velocities of helium exceed its ordinary diffusion velocities, caused by concentration gradient, around the reaction zone. This is clearly shown in Fig. 9.

The higher helium concentrations in reaction zone for the simulation with thermal diffusion result in much lower temperatures and lower acetylene concentrations in most of reaction zone. As a result, both inception and surface growth rates of the simulation with thermal diffusion are much lower than those of the simulation without thermal diffusion are. Therefore we can conclude that thermal diffusion has a significant effect on soot formation process in helium diluted ethylene/air diffusion flame, and thus should be taken into account in the numerical simulation.

Flame with 30% Argon Addition to Fuel

Figure 10 depicts the influence of thermal diffusion on integrated soot volume fraction for the flame with 30% argon addition to the fuel. It reveals that the influence of thermal diffusion on soot formation in this flame is similar to that in pure ethylene/air flame. The peak integrated soot volume fraction is overpredicted by 3.2% when thermal diffusion is neglected.

Thermal diffusion factors of argon are very small, with peak value less than 0.5, as shown in Fig. 11. Therefore although argon is driven to cold region due to the positive thermal diffusion factors, this effect is very small. As a result, the concentrations of argon in the reaction zone of the simulation with thermal diffusion are only slightly lower than those of the simulation without thermal diffusion. The influence of thermal diffusion on soot formation in this flame is mainly caused by the variations of concentrations of other species, such as those discussed for pure ethylene/air flame.

Flame with 30% Argon Addition to Air

The integrated soot volume fractions of the flame with 30% argon addition to air are plotted in Fig. 12. Again soot volume fraction is overpredicted when thermal diffusion is neglected. The peak integrated soot volume fraction is about 3.6% higher.

Similar to the addition of argon to fuel, although the concentrations of argon in the reaction zone are raised, temperatures are still higher when thermal diffusion is not accounted for. This is because that the thermal diffusion factors of argon are not positively high enough. The moving of other species caused by thermal diffusion and the nonlinear coupling effect result in higher temperatures in the reaction zone when thermal diffusion is neglected. Therefore soot volume fraction is overpredicted if thermal diffusion is neglected in the simulation of this flame.

The above numerical simulations show that thermal diffusion, caused by temperature gradient, does affect the prediction of soot formation in ethylene/air diffusion flame. Especially the influence is significant when there are some lighter chemical species, such as helium, existing in fuel or air stream. Therefore it is necessary to consider the effect of thermal diffusion in the modelling of soot formation in ethylene/air diffusion flames.

CONCLUSIONS

The influence of thermal diffusion on soot formation in coflow laminar ethylene/air and argon or helium diluted ethylene/air diffusion flames has been numerically investigated. The results show that thermal diffusion does affect the prediction of soot formation in laminar ethylene/air and argon or helium diluted ethylene/air diffusion flames. Soot volume fraction is overpredicted if thermal diffusion is not taken into account in the numerical simulation. Although the influence of thermal diffusion on soot formation is not very significant for pure ethylene/air and argon diluted ethylene/air diffusion flames, it becomes significant for helium diluted ethylene/air diffusion flame. Especially when helium was added to air of the flame, the peak soot volume fraction can be overpredicted by as high as 100%. The thermal diffusion velocity of helium in the flame can exceed the ordinary diffusion velocity.

REFERENCES

1. Dixon-Lewis, G., *Proc. Roy. Soc. London A307*: 111-135 (1968).
2. Greenberg, J.B., *Combust. Sci. Tech.* 24: 83-88 (1980).
3. Warnatz, J., *Numerical Methods in Laminar Flame Propagation* (ed by N. Peters and J. Warnatz), Braunschweig: Vieweg, pp.87-111, 1982.
4. Hancock, R.D., Schauer, F.R., Lucht, R.P., Katta, V.R., and Hsu, K.Y., *Proc. Comb. Inst.* 26: 1087-1093 (1996).
5. Ern, A. and Giovangigli, V., *Combust. Theory Modelling* 2: 349-372 (1998).
6. Ern, A., and Giovangigli, V., *Combust. Sci. Tech.* 149: 157-181 (1999).
7. Williams, B.A., *Combust. Flame* 124: 330-333 (2001).
8. McEnally, C.S., Schaffer, A.M., Long, M.B., Pfefferle, L.D., Smooke, M.D., Colket, M.B., and Hall, R.J., *Proc. Comb. Inst.* 27: 1497-1505 (1998).
9. Smooke, M.D., McEnally, C.S., Pfefferle, L.D., Hall, R.J., and Colket, M.B., *Combust. Flame* 117: 117-139 (1999).
10. Bennett, B.A., McEnally, C.S., Pfefferle, L.D., and Smooke, M.D., *Combust. Flame* 127:2004-2022 (2001).
11. Kennedy, I.M., Yam, C., Rapp, D.C., and Santoro, R.J., *Combust. Flame* 107: 368-382 (1996).
12. Gülder, Ö.L., Snelling, D.R., and Sawchuk, R.A., *Proc. Comb. Inst.* 26:2351-2358 (1996).
13. Guo, H., Liu, F., Smallwood, G.J., and Gülder, Ö.L., *Combust. Theory Modelling* 6: 173-187 (2002).
14. Liu, F., Smallwood, G. J., and Gülder, Ö.L., *AIAA paper* 99-3679.
15. Kee., R.J., Warnatz, J., Coltrin, M.E., and Miller, J.A., Sandia Report, *SAND 86-8246*.
16. Leung, K.M, Lindstedt, R.P., and Jones, W.P., *Combust. Flame* 87: 289-305 (1991).
17. Fairwhether, M., Jones, W.P. and Lindstedt, *Combust. Flame* 89:45 (1992).
18. Patankar, S.V., *Numerical Heat Transfer and Fluid Flow*, Hemisphere, New York, 1980.
19. Liu, Z., Liao, C., Liu, C. and McCormick, S., *AIAA* 95-0205.
20. Gregory P. Smith, David M. Golden, Michael Frenklach, Nigel W. Moriarty, Boris Eiteneer, Mikhail Goldenberg, C. Thomas Bowman, Ronald K. Hanson, Soonho Song, William C. Gardiner, Jr., Vitali V. Lissianski, and Zhiwei Qin http://www.me.berkeley.edu/gri_mech/.
21. Kee., R.J., Miller, J.A., and Jefferson, T.H., Sandia Report, *SAND 80-8003*.