

Rate-Controlled Constrained-Equilibrium Theory Applied to the Expansion of Combustion Products in the Power Stroke of an Internal Combustion Engine

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Abstract

Rate-controlled constrained-equilibrium method, firmly based on the second law of thermodynamics, is applied to the expansion of combustion products of methane during the power stroke of an internal combustion engine. The constraints used in this study are the elemental oxygen, hydrogen, carbon and nitrogen together with other four dynamic constraints of total number of moles, moles of DCO (CO+HCO), moles of free valence and moles of free oxygen. Since at chemical equilibrium, the mixture composition is dominated by H/O, CO/CO₂, and a few other carbon-containing species, almost independent of the fuel molecule, the set results in accurate predictions of the kinetic effects observed in all H/O and CO/CO₂ compounds and temperature history. It is shown that the constrained-equilibrium predictions of all the species composed of the specified atomic elements can be obtained independent of a kinetic path, provided their Gibbs free energies are known.

Keywords: Second law of thermodynamics, rate-controlled constrained-equilibrium (RCCE) method, constraints, chemical kinetics, internal combustion engine, power stroke.

1. Introduction

Equilibrium gas dynamics is based on the assumption that when a system undergoes either a heat or work interaction with the surrounding environment, the internal molecular relaxation processes are faster than changes brought about in the system's thermodynamic states due to interaction. Under this assumption the condition of local thermodynamic equilibrium (L.T.E.) is valid. L.T.E. leads to thermal equilibrium among various molecular degrees of freedom, enabling the definition of a single temperature. However, this is not generally the case, as energy redistribution among internal degrees of freedom (translation, rotation, vibration, and electronics) requires definite lengths of time, known as relaxation times (Vincenti, Kruger, 1965). If the interaction occurs on a time scale shorter or comparable with molecular relaxation, the internal dynamics of the system lags behind in re-establishing local thermodynamic equilibrium and the slow degree of freedom has to be treated by means of non-equilibrium thermodynamics.

This study is focused on gas phase chemical relaxation, where a chemically reacting system undergoes work interaction with the environment and is initially in a chemical equilibrium state. As pointed out by Keck (1990), an equilibrium state is meaningful only when the constraints, subject to which such a state is attained, are carefully determined and all equilibrium states are in fact constrained equilibrium states. At temperatures of interest to combustion, nuclear and ionization reactions can be assumed frozen and the fundamental constraints imposed on the system are conservation of neutral atoms. The cascade of constraints can be easily extended based on the existence of classes of slow chemical or energy-exchange reactions, which if completely inhibited would prevent the relaxation of the system to complete equilibrium. For

instance, total number of moles in a reacting system does not change unless a three body reaction occurs, radicals are not generated in the absence of chain branching reactions and the definition of a single temperature in a chemically reacting system is based on the observation that thermal equilibration among translation, rotation and vibration is in general faster than chemical reactions.

Based on the most profound law of nature, i.e. second law of thermodynamics, Rate-Controlled Constrained-Equilibrium (RCCE) method was originally developed by Keck and Gillespie (1971) and later by Keck and coworkers (Bishnu, Hamiroune, Metghalchi, Keck, 1997 and Hamiroune, Bishnu, Metghalchi, Keck, 1998), Tang and Pope (2004) and Jones and Rigopoulos (2005) to estimate the state of a nonequilibrium system by maximizing entropy at any time during the nonequilibrium evolution subject to the known constraints imposed on the system. The dynamics of the unrepresented part of the system is then determined by the requirement of constrained equilibrium.

According to the fundamental premise of RCCE, slow reactions in a complex reacting system impose constraints on its composition, which control the rate at which it relaxes to chemical equilibrium, while the fast reactions equilibrate the system subject to the constraints imposed by the slow reactions. Consequently, the system relaxes to chemical equilibrium through a sequence of constrained-equilibrium states at a rate controlled by the slowly changing constraints (Keck, 1990).

Morr and Heywood (1974) considered the problem of sudden cooling of the combustion products of aviation kerosene by passing the combustion products through an energy exchanger at constant pressure as a model for a gas turbine. They compiled a model for CO oxidation chemistry based on the fact that three body and CO oxidizing

reactions are generally slower than other reactions and confirmed the notion through comparison against experimental data. In this paper we look at the expansion of (CH₄/O₂/N₂) combustion products in the power stroke of an internal combustion engine, when N₂ is assumed to be inert, using the RCCE method. This method enables a systematic analysis of the underlying kinetics through testing different constraints with the least amount of effort.

2. Physical Model

The physical model is shown in Figure 1. It is assumed that the power stroke begins with the combustion products at a complete chemical equilibrium state and that expansion occurs so rapidly that heat conduction does not occur through the piston wall. It is further assumed that the gas composition is homogeneous at any time during expansion. Volume is assumed to be a prescribed function of time (Heywood, 1988):

$$\frac{V(t)}{V_c} = 1 + \frac{1}{2}(r_c - 1) \times f(\theta)$$

$$f(\theta) = [R + 1 - \cos \theta(t) - \sqrt{R^2 - \sin^2 \theta(t)}]$$

$$R = \frac{l}{a} \quad (1)$$

where $V(t)$, V_c , r_c and θ represent the instantaneous volume of the cylinder, clearance volume, compression ratio and the instantaneous crank angle respectively. The numerical values used in this study are $V_c = 125 \text{ mlit}$, $r_c = 11$, and $R=3$. The power stroke is also identified by $0 \leq \theta \leq 180$.

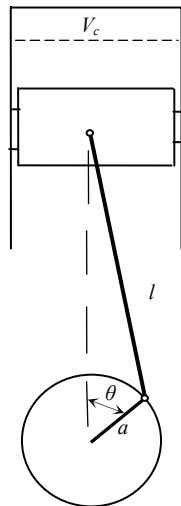


Figure 1. Schematic of the physical model.

3. Governing Equations in RCCE Form

The detailed formulation of RCCE in constrained-potential form can be found in earlier works (Janbozorgi, Gao, Metghalchi, Keck, 2006), so we avoid repeating the procedure in detail here and address the important points. Consistent with the perfect gas assumption, the constraints imposed on the system by the reactions are assumed to be a linear combination of the mole number of the species present in the system,

$$C_i = \sum_{j=1}^{N_s} a_{ij} N_j, \quad i = 1, \dots, N_c \quad (2)$$

The constrained-equilibrium composition of a system found by maximizing the entropy or minimizing the Gibbs free energy subject to a set of constraints using the method of Lagrange multipliers is (Keck, 1990):

$$N_j = Q_j \exp\left(-\sum_{i=1}^{nc} a_{ij} \gamma_i\right), \quad j = 1, \dots, N_s \quad (3)$$

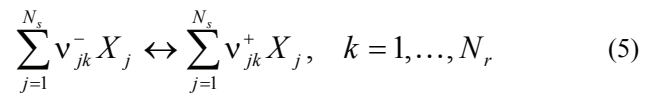
where,

$$Q_j = \frac{p_c V}{RT} \exp(-\mu_j^0) \quad (3a)$$

Clearly, the partition function of each species depends on temperature, volume and the standard Gibbs free energy of the species. The time rate of change of constraints or equivalently, constraint potentials, can be obtained by taking the time derivative of Eqn (2) as:

$$\dot{C}_i = \sum_{j=1}^{N_s} a_{ij} \dot{N}_j \quad (4)$$

It is assumed that changes in the chemical composition are the results of chemical reactions of the form



The phenomenological expression of chemical kinetics can be used to replace \dot{N}_j in equation (4)

$$\dot{N}_j = \sum_{k=1}^{N_r} \nu_{jk} r_k \quad (6)$$

where

$$r_k = r_k^+ - r_k^- \quad (6-a)$$

$$\nu_{jk} = \nu_{jk}^- - \nu_{jk}^+ \quad (6-b)$$

Substituting Eqn (6) into (4) results in the required time rate of change of the constraint C_i :

$$\dot{C}_i = \sum_{k=1}^{N_r} b_{ik} r_k \quad (7)$$

$$b_{ik} = \sum_{j=1}^{N_s} a_{ij} \nu_{jk} \quad (7-a)$$

Clearly, a reaction k for which the value of b_{ik} is zero for all constraints i is in constrained equilibrium. All reactions in the mechanism must satisfy this condition for elemental constraints. In the absence of any spatial nonhomogeneity, the implicit differential equations governing the constraint potentials can be obtained by substituting Eqn (7) and the

time derivative of Eqn (3) into Eqn (4), the final result of which is:

$$\sum_{n=1}^{N_c} C_{i\gamma} \dot{\gamma}_n - C_{iV} \frac{\dot{V}}{V} - C_{iT} \frac{\dot{T}}{T} + \sum_{k=1}^{N_r} b_{ik} r_k = 0 \quad (8)$$

where

$$C_{i\gamma} = \sum_{j=1}^{N_s} a_{ij} a_{nj} [N_j] \quad (8-a)$$

$$C_{iV} = \sum_{j=1}^{N_s} a_{ij} [N_j] \quad (8-b)$$

$$C_{iT} = \sum_{j=1}^{N_s} a_{ij} \frac{E_j}{RT} [N_j] \quad (8-c)$$

In cases where state variables other than T and V are used, additional equations for these are required. In the present work, the energy of the system

$$E = \sum_{j=1}^{N_s} E_j N_j \quad (9)$$

is used to replace T . The required energy equation can be obtained from the first law of thermodynamics. As stated in Section 3, we assume that expansion occurs so rapidly that heat conduction through the piston wall is frozen and that the work term is due only to volume change. Differentiating Eqn (9) with respect to time and using the differential form of Eqn (3) results in the required energy equation:

$$\sum_{n=1}^{N_c} D_{E\gamma} \dot{\gamma}_n + D_{EV} \frac{\dot{V}}{V} + D_{ET} \frac{\dot{T}}{T} - \dot{E} = 0 \quad (10)$$

where

$$D_{E\gamma} = - \sum_{j=1}^{N_s} E_j a_{nj} [N_j] \quad (10-a)$$

$$D_{EV} = \sum_{j=1}^{N_s} E_j [N_j] \quad (10-b)$$

$$D_{ET} = \sum_{j=1}^{N_s} \left(c_{vj} T + \frac{E_j^2}{RT} \right) [N_j] \quad (10-c)$$

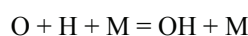
$$\dot{E} = -p\dot{V} \quad (10-d)$$

Upon solving the differential equations for the γ_i vector, the constrained-equilibrium composition of the system can be found at each time step from Eqn (3). Of particular importance at this point is to notice that every species for which the Gibbs free energy is known can be included in the model, *even though they may not explicitly participate in the kinetic mechanism*, and an estimation of its dynamic evolution can be obtained based on the value of the known constraints. Such dynamic evolution is dictated by the requirement of constrained-equilibrium. In the limit of local chemical equilibrium, where all reactions are in constrained-equilibrium, the constraints imposed by the conservation of neutral atoms enables determining the concentration of every possible species made up of the same elements. Such an approach is the basis of the widely used STANJAN and NASA equilibrium codes (Reynolds, Gordon, McBride, 1971). Evidently, different sets of constraints can be handled with the same ease using the above formulations.

4. Constraints

The selection of appropriate constraints is the key to the successful application of the RCCE method. Among the general requirements for the constraints are that they must (1) be linearly independent combinations of the species mole numbers, (2) include the elements, (3) determine the energy and entropy of the system within experimental accuracy, and (4) hold the system in the specified initial state. In addition, they should reflect whatever information is available about rate-limiting reactions controlling the time evolution of the system. The constraints used in this study are listed in Table 1.

According to the Le Châtelier principle, when a highly dissociated mixture undergoes an interaction with the environment which lowers the gas temperature and density, the internal dynamics shift in the exothermic direction to minimize the cooling effect of interaction. As a result, three body recombination reactions become an important part of the energy restoration process. The total number of particles in the system does not change unless one such reaction occurs. This is the rationale for putting a constraint on total number of moles; M . Among the reactions changing this constraint are:



Oxidation of CO to CO₂ constitutes an important part of the exothermic relaxation which is dominated by several reactions, such as:

Table 1. Constraints Used In This Study

1	EN	Elemental nitrogen
2	EC	Elemental carbon
3	EO	Elemental oxygen
4	EH	Elemental hydrogen
5	M	Total number of moles
6	DCO	Moles of HCO+CO
7	FV	Moles of free valence (any unpaired valence electron)
8	FO	Moles of free oxygen (any oxygen not directly bound to another oxygen)

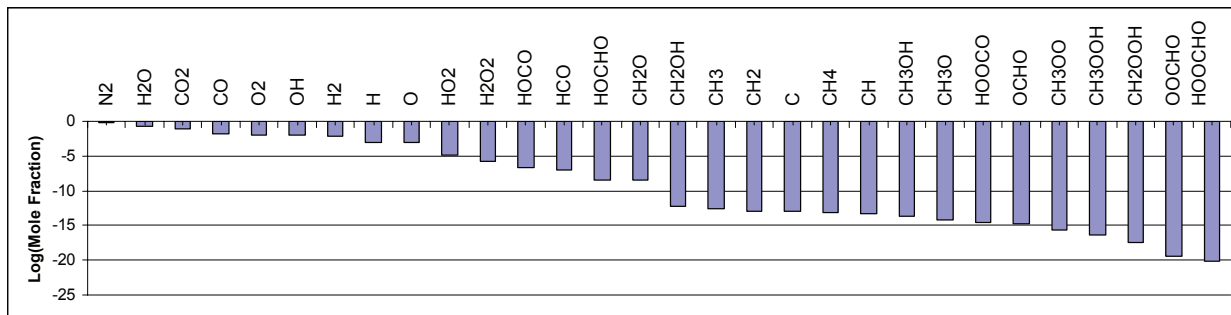


Figure 2. Chemical equilibrium composition of stoichiometric $CH_4/O_2/N_2$ mixture.



(R1) is a generic reaction that involves interaction with the radical pool, which is in this case dominated by H/O radicals, and is generally faster than others because it involves small or zero activation energy. In order to allow these reactions to be in constrained equilibrium, a constraint on HCO+CO is introduced. This constraint further controls the exothermic path from CO to CO₂. The energy restoration process also requires chain branching reactions that act to produce more OH radicals, which itself feeds exothermic water and CO₂ formation reactions. This condition requires constraints on both free valance, FV, and free oxygen, FO. The most important reaction belonging to these constraints is



5. Results and Discussion

Equations (8) and (10) were integrated using DASSL (Petzold 1982). A mixture of ($CH_4/O_2/N_2$) is compressed to an initial temperature of 735 K and initial pressure of 15 atm. It is further assumed that compression occurs so rapidly that chemical reactions are frozen. The power stroke is then assumed to start with the combustion products, corresponding to instantaneous conversion of fuel/oxidizer under a constant-volume, constant energy combustion process at the clearance volume. Initial conditions for expansion can be obtained from STANJAN. The chemical equilibrium composition for all 30 species corresponding to an initially stoichiometric mixture of ($CH_4/O_2/N_2$) is shown in Figure 2. The equilibrium temperature and pressure are $T_{eq} = 2869$ K and $P_{eq} = 59.5$ atm. Clearly, it is dominated by H/O and CO/CO₂ components followed by HOCO, HCO, CH₂O, and HOCHO as the next important carbon-containing compounds. Such an equilibrium mixture is almost fuel independent.

From a kinetics standpoint, three body reactions have small or zero activation energies, making them almost temperature insensitive and rather highly pressure (density) sensitive, whereas the rate of bimolecular reactions which involve activation energies are temperature sensitive. Therefore, sudden cooling to low temperatures and lowering density depress the rate of recombination and exothermic bimolecular reactions markedly and the exothermic processes lag in their attempt to restore the equilibrium. A failure to release latent energy of molecule formation enhances the cooling and puts the system farther

out of equilibrium. If expansion is fast enough, the exothermic lag grows indefinitely and the composition becomes frozen.

Figures 3 and 4 represent the frozen, local thermodynamic equilibrium (L.T.E.) and nonequilibrium temperature and CO profiles during expansion of combustion products of an initially stoichiometric mixture of ($CH_4/O_2/N_2$). It should be mentioned that since the mixture is assumed to be spatially homogeneous at each instant of time, local chemical equilibrium is equivalent with L.T.E. Physically, frozen chemistry corresponds to the case in which expansion occurs so rapidly that chemistry is completely frozen, whereas L.T.E. is the case when expansion occurs extremely slowly so that all the kinetic processes are instantaneously equilibrated. L.T.E. calculations are carried out using the general formulations but using only elemental constraints. In both cases the expansion is isentropic but in the former the species are the fixed constraints, whereas in the later neutral atoms are the fixed constraints and the chemical composition of the system changes according to the requirement of L.T.E. in response to changes in the state variables.

Obviously, L.T.E. calculations result in the highest temperature and highest conversion of CO to CO₂. The opposite is true for the frozen chemistry. Predictions of the finite-rate chemistry are bounded by these two extremes. The results of such predictions using a mechanism comprising 133 reactions and 30 species, designated by "detailed kinetics", are compared with RCCE results when constraints are added one at a time. All RCCE results include elemental constraints. Thus labels refer to constraints in addition to elementals.

The slowest constraint is imposed by three-body recombination/dissociation reactions. The effect of all such reactions is captured by putting a constraint on the total number of moles (M).

Evidently adding to the list a constraint on $DCO = HCO + CO$ does not change the predictions noticeably, meaning that these reactions (reactions similar to R3 and R4) are nearly in constrained equilibrium. However, the collective effect of DCO, FV, and FO results in perfect match between RCCE using 8 constraints and detailed kinetics using 30 species.

It is also interesting to notice that the state of the gas follows exactly the predictions of L.T.E. during the early stage of expansion, when the piston speed is very slow. Departures from local thermodynamic equilibrium emerge as the piston speed increases.

In order to check the validity of the identified constraints under faster expansions, the engine speed is increased to 6,000 rpm and 12,000 rpm. In this case the kinetic effects are only observed in species profiles and not

in temperature. Figures 5 and 6 demonstrate these observations. Figures 7 and 8 show the same set of results for expansion of combustion products of a mixture of ($\text{CH}_4/\text{O}_2/\text{N}_2$) with an initial equivalence ratio of 0.8. Progressive improvements of the results by adding constraints one at a time is evident. Pressure at the end of

the stroke is 3 atm. Figures 9 and 10 show the same result for the case in which the initial equivalence ratio is 1.2. Contrary to the results corresponding to a stoichiometric mixture, Figure 4, in this case three body reactions are in constrained equilibrium and DCO-changing reactions are rate limiting.

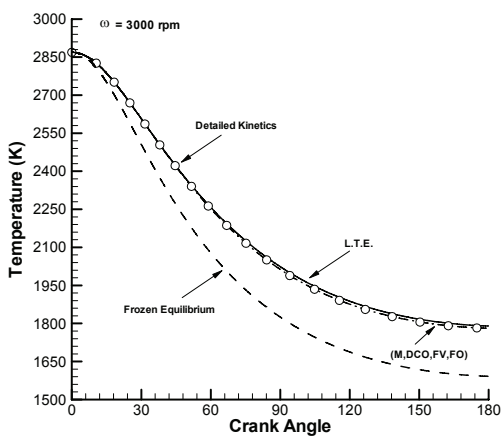


Figure 3: RCCE prediction of temperature profile compared with the predictions of detailed kinetics at an engine speed of 3000 rpm and initial pressure of $P_i=59.5$ atm.

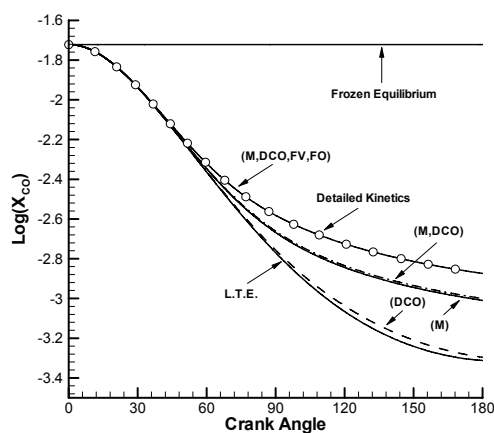


Figure 4: Constraint-dependence of CO profiles compared with the predictions of detailed kinetics at an engine speed of 3000 rpm and initial pressure of $P_i=59.5$ atm and initial temperature of $T_i=2869$ K

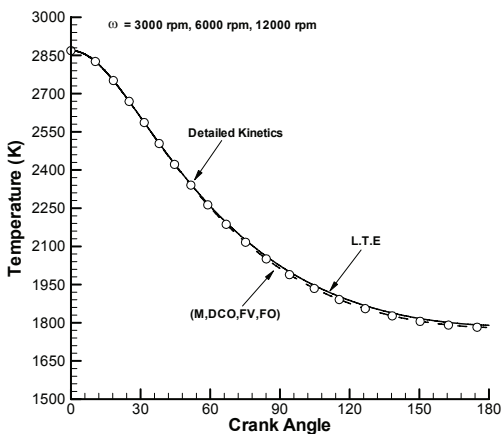


Figure 5: Engine-speed dependence of temperature profiles compared with the predictions of detailed kinetics at the equivalence ratio of 1.0.

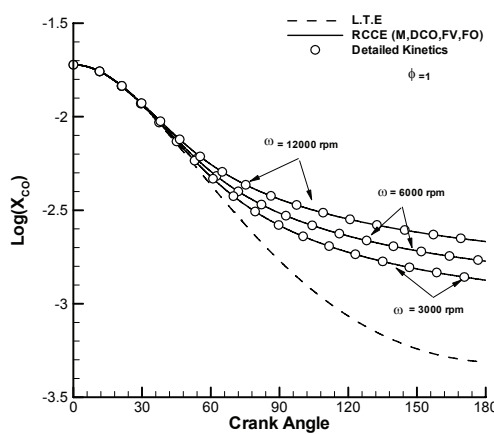


Figure 6: Engine-speed dependence of CO profiles compared with the predictions of detailed kinetics at the equivalence ratio of 1.0.

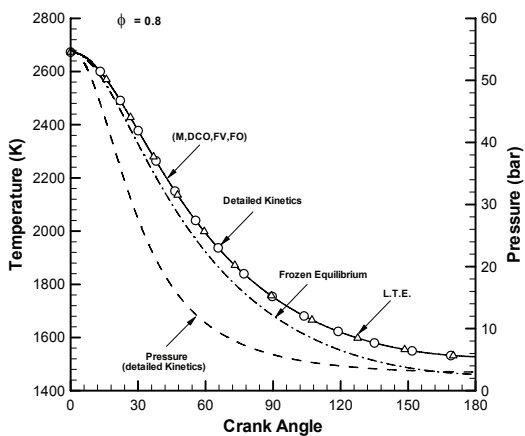


Figure 7: Temperature and pressure profiles under different conditions at an engine speed of 3000 rpm, initial temperature of $T_i=2673$ K, initial pressure of $P_i=54.83$ atm and equivalence ratio of 0.8.

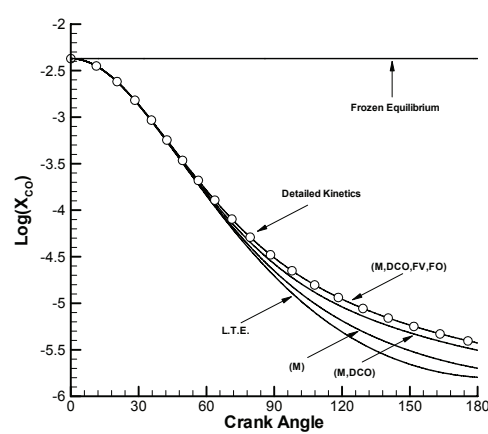


Figure 8: CO profiles under different sets of constraints at an engine speed of 3000 rpm, initial temperature of $T_i=2673$ K, initial pressure of $P_i=54.83$ atm and equivalence ratio of 0.8.

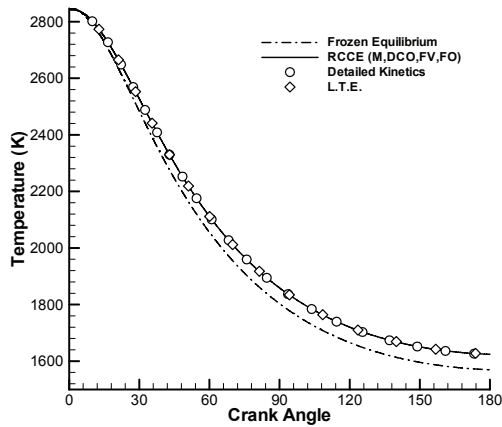


Figure 9. Temperature profiles under different sets of constraints at an engine speed of 3000 rpm, initial temperature of $T_i=2846.8$ K, initial pressure of $P_i=60.5$ atm and equivalence ratio of 1.2.

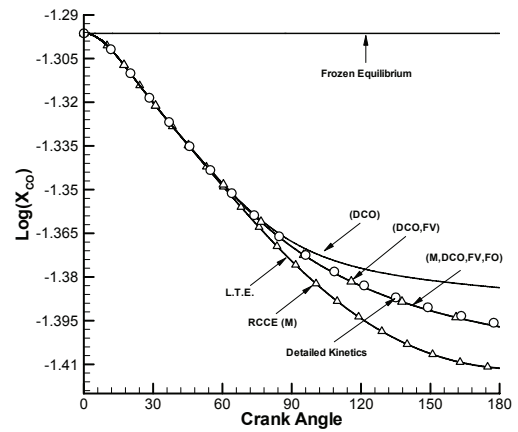
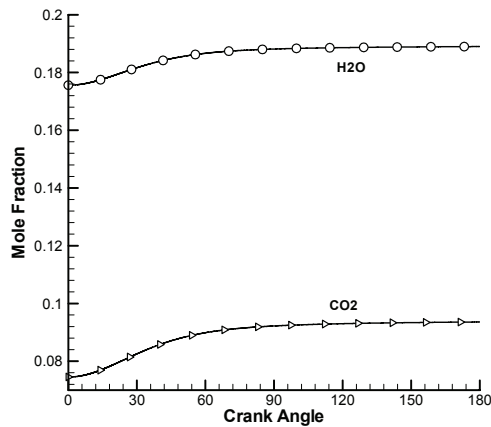


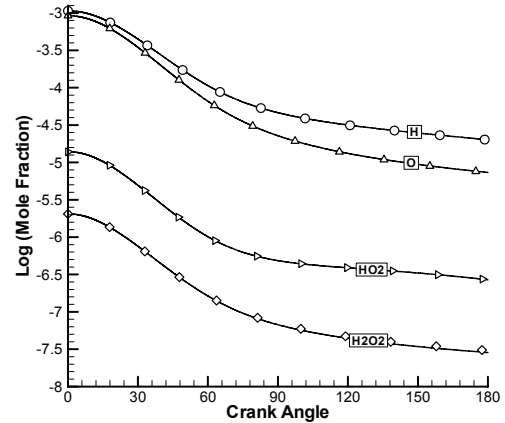
Figure 10. CO profiles under different sets of constraints at an engine speed of 3000 rpm, initial temperature of $T_i=2846.8$ K, initial pressure of $P_i=60.5$ atm and equivalence ratio of 1.2.

Figure 11(a,b,c,d) represents the profiles of H_2O , CO_2 , H , O , HO_2 , H_2O_2 , $HOCO$, HCO , $HOCHO$, CH_2O , CO , O_2 , H_2 , OH corresponding to an equivalence ratio of 1.0 at an engine speed of 3000 rpm. Comparison has been made between RCCE calculations using 8 constraints against detailed kinetics using 30 species and 133 reactions. It should be noticed that $HOCHO$, CH_2O and $HOCO$ are not directly constrained and their evolution is determined based on the requirement of constrained equilibrium. This demonstrates one of the most important features of RCCE

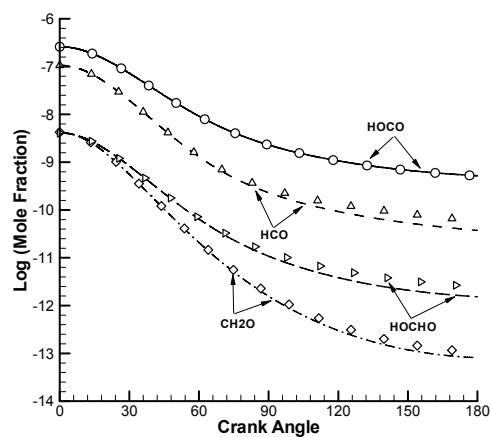
that with a few constraints the concentrations of all possible species composed of the specified elements can be readily obtained, provided their Gibbs free energies are known. Having defined the constraint $DCO=HCO+CO$, the concentration of HCO is also determined based on an internal equilibrium with CO . Clearly, the single constraint DCO is able to give quite acceptable predictions for CO , Figure 11d, and HCO , Figure 11c, which confirms the partial equilibrium between them.



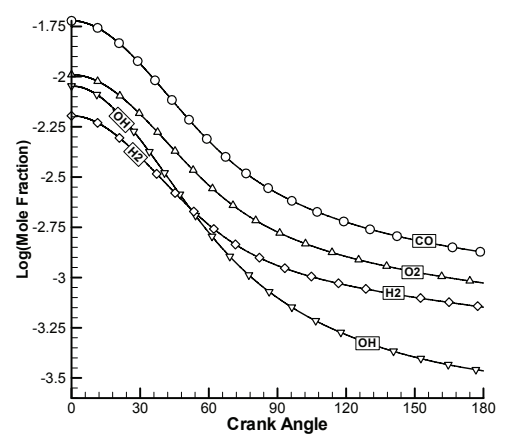
(a)



(b)



(c)



(d)

Figure 11. Profile of different species for $T_i=2869$ K, $P_i = 59.5$ atm at $\phi = 1$ and $\omega = 3000$ rpm.

6. Concluding Remarks

Local thermodynamic equilibrium (L.T.E.), frozen chemistry, detailed kinetics and Rate-controlled constrained-equilibrium calculations of the expansion of (CH₄/O₂/N₂) combustion products during the power stroke of an internal combustion engine were conducted. Looking at the dynamics of the expansion process, a set of 8 generalized constraints was identified. The set gives perfect matches with the results of detailed kinetics. The constraints identified are quite general and are able to handle relaxation from an initial equilibrium state of the combustion product of any hydrocarbon fuels. Under stoichiometric conditions, DCO-changing reactions are in constrained equilibrium and M-changing reactions are rate limiting, whereas, making the mixture richer, puts M-changing reactions in equilibrium and DCO-changing reactions out of equilibrium.

Of the salient features of the RCCE technique is that it is based on the most profound law of thermodynamics, Maximum Entropy Principle. Every species made up of the specified elements can be included in the model, provided its Gibbs free energy is known. In the absence of any direct constraint on a species or group of species, they will evolve according to the requirement of constrained-equilibrium. RCCE has the dazzling ability to carry out local thermodynamic equilibrium, frozen equilibrium and rate-controlled constrained-equilibrium calculations with the same ease, by implementing different classes of constraints. In the limit where the number of constraints equals the number of species, the method becomes exact.

7. Acknowledgments

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8. Nomenclature

C_i	Value of constraint i
a_{ij}	Value of constraint i in species j
N_j	Number of moles of species j
$[N_j]$	Concentration of species j
Q_j	Partition function of species j
γ_i	Constraint potential (Lagrange multiplier) conjugate to constraint i
μ_j°	Nondimensional standard Gibbs free energy of species j , $(h_j^\circ - Ts_j^\circ)/RT$
E_j	Molar energy of species j
b_{ik}	Change in constraint i by reaction k
r_k	Net rate of reaction k
r_k^+	Forward rate of reaction k
r_k^-	Reverse rate of reaction k
v_{ij}^+	Stoichiometric coefficient of product species j in reaction i
v_{ij}^-	Stoichiometric coefficient of reactant species j in reaction i

N_s	Number of species
N_c	Number of constraints
N_r	Number of reactions
c_{vj}	Frozen molar heat capacity of species j at constant volume
T	Temperature
V	Volume
P	Pressure
p°	Standard (atmospheric) pressure
E	Total energy
R	Universal gas constant

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