Simulation of turbulent mixing and chemical reaction in a Partially Stirred Reactor using the Direct Quadrature Method of Moments.

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Abstract. In this study we investigate the use of Direct Quadrature Method of Moments for simulating turbulent mixing and chemical reaction in a partially stirred reactor (PaSR). A partially stirred reactor is characterized by perfect macromixing but imperfect micromixing. PaSRs are the building blocks of zone models for industrial scale chemical reactors, but they can also be considered to be a single computational grid in a more detailed CFD simulation such as the LES-FMDF. We consider a two stream mixing problem of a hydrocarbon fuel and air with an Arrhenius type chemical reaction rate that is commonly used for combustion processes. We consider a single step, a two step competitive-consecutive, and a more detailed chemical reaction mechanism. For the mixing, we use a common turbulent mixing model called the Interaction by Exchange with Mean (IEM). We then compare the solutions obtained using DQMOM with solutions obtained using Monte Carlo simulations. Due to the exponential dependence of the chemical reaction rate, the equations for the moments are unclosed and there is some error in the DQMOM solutions. Further, as the dimension of the pdf increases (for example by considering more complicated reaction mechanisms involving larger number of species), moment methods become less efficient and difficult to solve. However for simplified chemical schemes and in use with CFD models, DQMOM appears to be a very promising tool. The objective of the comparison of the Monte Carlo and DQMOM solutions is to investigate the tradeoffs inherent in both methods and attempt to provide guidelines as to when one method may be preferred over another.
Keywords: Partially Stirred Reactor, Turbulent Mixing, Chemical Reactions, Combustion, Direct Quadrature Method of Moments, Monte Carlo Simulations.

The prediction of mixing and chemical reactions in the presence of a turbulent flow field is of great importance in the chemical process industries and has been the subject of intense research. In a turbulent flow chemical reactor there are fluctuations in the flow field and the chemical species concentrations over a very wide range of scales. A computational simulation of an industrial scale chemical reactor that resolves the fluctuations of the velocity field and chemical species down to the smallest time and length scales is intractable with the currently available software and hardware. While more detailed simulations of the flow field have become possible due to advances in the techniques of computational fluid dynamics (an example is the method of Large Eddy Simulation (LES)), the extension of these methods to the simulation of reacting scalars within a turbulent flow field is not straightforward. A basic premise of methods like the LES is that only the larger energy containing velocity fluctuations need to be resolved while the smallest scales in a turbulent flow are universal and can be described by known statistics. However, the mixing of scalars and hence the chemical reactions take place in the molecular scales and these processes are certainly not universal, i.e. they depend on the details of how the reactants are fed into the vessel, the rate of stirring and the large scale flow field as well as the smaller scale velocity fluctuations that enhance mixing.

In the past, chemical reactors were studied in two limiting cases: fast reactions and fast mixing. For fast reactions, a mixture fraction approach is used to describe mixing and the conversion of reactants to products is assumed to be instantaneous. For chemical reactions that are slow compared to all the other flow-related processes, the details of the flow field are relatively unimportant and the flow in the reactor can be characterized as plug flow or as continuously stirred. The only effect of the flow field on chemical reactions is through the residence time (see for instance textbooks such as Hill (1977)). In between there is a large class of problems in which both the mixing and the chemical reactions have to be accounted for. An example is the extinction and reignition phenomena in combustion processes. The complex nature of these problems can be appreciated when one observes that chemical reactions proceed through a large number of intermediate steps involving a large number of chemical species. Further, scalar dispersion is caused by turbulent velocity fluctuations, and ultimately the mixing is governed by molecular processes. Since it is infeasible to resolve molecular scale processes in a computational simulation of the flow process, stochastic methods are used for describing molecular mixing and reaction. In computational terminology, these processes are referred to as “subgrid-scale” processes meaning that these processes occur in length and time scales smaller than computational grid resolution and therefore it is only possible to obtain statistical information on how these processes evolve. Stochastic methods involve the solution of a pdf transport equation. The pdf usually carries information about species concentrations and temperatures and evolves in a higher dimensional space. Hence its solution requires alternative methods like Monte Carlo simulations or moment methods. A detailed account of the pdf transport method for turbulent reacting flows is given in Pope (1985). Needless to say the computational requirements for a using a statistical method such as Monte Carlo with some grid based simulation is very large. As an example, in a simulation of a jet diffusion flame, Branley and Jones (2001) used 624,100 computational cells. To represent the subgrid scale processes using Monte Carlo simulations, around 20-100 particles are required in each grid cell to adequately represent a pdf. Then differential equations need to be solved for each of the particles.

Before going into details it is helpful to consider the bigger picture of the problem. When simulating a stochastic process describing mixing and chemical reaction, we are essentially tracking all the possible outcomes of the process and probabilities associated with each outcome. The information required to describe a mixing and reacting system at each instant can be represented by a data matrix as shown in figure 1. This matrix consists of m rows of samples that represent all the possible states of the system. The samples are also referred to as particles. The n columns of the matrix define the information carried by each particle. Graphically they are a swarm of m points in n-dimensional space. The total number of columns gives the dimension of the system which is the number of independent quantities required to describe the system. Chemical reactions are notoriously high dimensional systems. For instance a detailed mechanism for the oxidation of methane involves around 40 species and 200 reactions. In this case the data matrix consists of 40 columns. Major research in the simplification of chemical kinetics involves reducing n, the dimension of the system. Chemical mechanism reduction schemes such as the ILDM (Maas and Pope (1992)) are techniques used for dimension reduction. The number of rows, m, are the number of samples required to accurately represent the underlying pdf. For a deterministic system such as a laminar combustion problem only a single sample is needed. In a turbulent combustion problem where there are fluctuations in species properties caused by fluctuations in the flow field, a large number of samples are required to obtain good statistics. For chemical reactions the particles do not interact but the proportions of the various species (i.e. the entries in the columns for a particular row) change continuously. In combustion applications one usually has to solve a stiff

Table 1. Schematic representation of a data matrix describing a reacting system.

	Dimensions (eg. species mass fractions)				
Samples	**Y_1**	**Y_2**	**……………**	**Y_{n-1}**	**Y_n**
N_1					
N_2					
	↑	**Statistical Reduction**	←	**Dimension Reduction**	
N_{m-1}					
N_m					

non-linear ordinary differential equation (ODE) to obtain the trajectory of each particle in the high dimensional space. This operation involves the greatest computational cost of a reacting flow simulation. Hence, for a fixed dimension, any technique that provides a systematic method of reducing the number of particles leads to significant savings in computation. Along with the reactions, molecular mixing also takes place. For particle systems the mixing needs to be modeled. Several mixing models have been proposed in the literature. The most straightforward is the Interaction by Exchange with the Mean (IEM) method in which the particles move deterministically toward the mean. Stochastic mixing models that involve interactions between particles have also been proposed. A more detailed mixing model that uses the concept of a minimum spanning tree also exists. The EMST model proposed by Subramaniam and Pope (1998) describes mixing due to interactions between “similar particles”, particles that are close in the n-dimensional space. In analogy to “dimension reduction”, “statistical reduction” is also possible, where the number of samples/particles m is reduced. In a sense, the ISAT technique invented by Pope (1997) can be considered to be a statistical reduction method in which samples that are close in composition space are grouped into clusters. As will be shown later moment methods are methods in which the sample size is reduced drastically. These methods provide the greatest statistical reduction possible. For instance, in the Direct Quadrature Method of Moments (DQMOM) technique (Fox (2003)), usually two or three quadrature points are used which in our example above is equivalent to using two or three particles.

In the rest of the paper we investigate the use of Direct Quadrature Method of Moments to simulate turbulent mixing and chemical reaction problems. To isolate the problem, mixing and chemical reaction in a partially stirred reactor (PaSR) model is considered. A PaSR is a reactor that is characterized by perfect macromixing (i.e. no spatial gradients of the scalars) but imperfect micromixing (i.e. mixing is not complete at the molecular scale). A PaSR can be considered to be a single computational grid in a more detailed CFD simulation. It can also be considered as an element of zone models for industrial scale chemical reactors. We intend to illustrate both the application of DQMOM and to evaluate its accuracy. For this reason, we consider a 1-step, 2-step, and detailed combustion reaction mechanisms involving 19 chemical species. These cases cover low dimensional as well as high dimensional cases described above. While for low dimensional systems, DQMOM gives excellent performance in terms of speed and accuracy for a number of problems (see for instance Marchisio and Fox (2003)), the evaluation of its performance in higher dimensions has not been investigated.

1. DQMOM model for a partially stirred reactor.

Let us consider an adiabatic, isobaric chemical reactor with two inlet streams. One stream carries the fuel and the other air. An outlet stream removes the products. Mixing and chemical reactions occur inside the reactor. The pdf describing the system is the joint scalar pdf $f(\vec{\psi}) = f(\phi_1, \phi_2, ..., \phi_d)$ where each ϕ_k is the mass fraction of the kth species or the temperature. The governing equation for the density weighted pdf $\tilde{f}(\vec{\psi}) = \frac{\rho(\vec{\psi}) f(\vec{\psi})}{\langle \rho(\vec{\psi}) \rangle}$ is (Ren and Pope (2004)).

$$\frac{\partial \tilde{f}}{\partial t} = -\frac{\tilde{f}}{\tau_{res}} + \frac{1}{\tau_{res}}\left[P\tilde{f}^{(O)} + (1-P)\tilde{f}^{(F)}\right] - \frac{\partial}{\partial \psi_\alpha \psi_b}\left[\left\langle \Gamma \frac{\partial \phi_\alpha}{\partial x_i}\frac{\partial \phi_\beta}{\partial x_i} \middle| \vec{\psi} \right\rangle \tilde{f}\right] - \frac{\partial}{\partial \psi_\alpha}\left[S_\alpha(\vec{\psi})\tilde{f}\right] \tag{1.1}$$

This equation is obtained by integrating the joint scalar pdf transport equation (Pope (1985)) assuming statistical homogeneity. As per Ren and Pope (2004), the residence time $\tau_{res} = \frac{m}{\dot{m}_{ox} + \dot{m}_{fu}}$ and $P = \frac{\dot{m}_{ox}}{\dot{m}_{ox} + \dot{m}_{fu}}$, where m is the mass of gas inside the reactor and $\dot{m}_{ox}, \dot{m}_{fu}$ are inlet mass flow rates of the oxidizer and fuel respectively. The equivalence ratio is then given by $\phi = \frac{(1-P)}{(1-P_{st})}$ where P_{st} is the value of P for which the inflow mixture gives stoichiometry. The third term in the left takes into account the molecular mixing and the fourth term the chemical reactions. The mixing term $\frac{\partial}{\partial \psi_\alpha \psi_b}\left[\left\langle \Gamma \frac{\partial \phi_\alpha}{\partial x_i}\frac{\partial \phi_\beta}{\partial x_i} \middle| \vec{\psi} \right\rangle \tilde{f}\right]$ involves a conditional expectation and needs to be modeled. In this study we use the IEM model with a mixing time scale given by τ_{mix} (Dopazo (1975)).

$$\frac{\partial}{\partial \psi_\alpha \psi_b}\left[\left\langle \Gamma \frac{\partial \phi_\alpha}{\partial x_i}\frac{\partial \phi_\beta}{\partial x_i} \middle| \vec{\psi} \right\rangle \tilde{f}\right] = \frac{\partial}{\partial \psi_\alpha}\left[\frac{\left(\phi_\alpha - \bar{\phi}_\alpha(t)\right) f}{\tau_{mix}}\right] \tag{1.2}$$

The inlet streams carry pure fuel (mass fraction ϕ_{fu}^0 and temperature ϕ_T^0) and pure air (mass fraction ϕ_{ox}^0 and temperature ϕ_T^0) given by :

$$\begin{aligned} \tilde{f}^{(F)} &= \delta(\phi_{fu} - \phi_{fu}^0)\delta(\phi_T - \phi_T^0) \prod_{\alpha \neq fu, T} \delta(\phi_\alpha - 0) \\ \tilde{f}^{(O)} &= \delta(\phi_{ox} - \phi_{ox}^0)\delta(\phi_T - \phi_T^0) \prod_{\alpha \neq ox, T} \delta(\phi_\alpha - 0) \end{aligned} \tag{1.3}$$

The initial condition is pure air inside the reactor.

$$\tilde{f}(t=0) = \tilde{f}^{(O)} \tag{1.4}$$

DQMOM is a presumed pdf method that works by forcing a number of moments of the pdf to evolve correctly. The details can be found in Fox (2003). In the DQMOM formulation the underlying pdf $\tilde{f}(\vec{\psi})$ is represented by a series of delta functions.

$$\tilde{f}(\vec{\psi}; t) = \sum_{n=1}^{N_Q} W_n(t) \prod_{k=1}^{d} \delta(\phi_k - \hat{\phi}_{k,n}(t)) \tag{1.5}$$

where $W_n(t)$ is the quadrature weight, $\hat{\phi}_{k,n}$ is the k$^{\text{th}}$ coordinate of the n$^{\text{th}}$ quadrature point. N_Q is the total number of quadrature points, and d is the dimension of the space. By substituting the presumed form (1.5) into equation (1.1) and using the methods detailed in Fox (2003), we get equations for the evolution of the weights $W_n(t)$ and the weighted quadrature points $S_{k,n}(t) = W_n(t)\hat{\phi}_{k,n}(t)$,

$$\frac{dW_n}{dt} = a_n ; n = 1,.., N_Q \qquad 1.6$$

$$\frac{\partial S_{k,n}}{\partial t} = b_{k,n} - \left[\left(\frac{\phi_{k,n} - \bar{\phi}_k}{\tau_{mix}}\right) + S_k(\vec{\psi}_n)\right] W_n ; n = 1,.., N_Q ; k = 1..d$$

The $N_Q(d+1)$ source terms a_n and $b_{k,n}$ are obtained by forcing $N_Q(d+1)$ moments of the pdf to evolve correctly. This essentially involves solving a matrix equation of the form $A\alpha = \beta$ where A is an $N_Q(d+1)$ X $N_Q(d+1)$ matrix for which each row is of the form

$$A = \left[\underbrace{\left(1 - \sum_{\alpha=1}^{d} m_\alpha\right) \prod_{k=1}^{d} \phi_{k,n}^{m_\alpha} ; n = 1,..., N_Q ;}_{(N_{Qt})cols} \underbrace{\frac{\partial}{\partial \phi_{\alpha,n}} \left(\prod_{k=1}^{d} \phi_{k,n}^{m_k} \right) ; n = 1,..., N_Q , \alpha = 1,..., d}_{(N_Q \times d)cols} \right], \qquad 1.7$$

and there are $N_Q(d+1)$ rows given by $N_Q(d+1)$ d-tuples $(m_1,..., m_d)$. This choice of A ensures that $N_Q(d+1)$ moments $M_{m_1,...,m_d}$ moments evolve consistently. The vector α contains the source terms $(a_n ; n = 1,..., N_Q ; b_{k,n} ; n = 1,..., N_Q ; k = 1,..., d)$ and β consists of terms due to mixing, reactions, inlets and outlet. Each of the $N_Q(d+1)$ rows of β determined by the d-tuple $\{m_\alpha\}$ is given by

$$\beta_{\{m_\alpha\}} = \left(\frac{1}{\tau_{res}}\right) \left[P\left(\phi_{ox}^0\right)^{m_{ox}} \left(\phi_T^0\right)^{m_T} \prod_{\alpha \neq ox,T} 0^{m_\alpha} + (1-P)\left(\phi_{fu}^0\right)^{m_{fux}} \left(\phi_T^0\right)^{m_T} \prod_{\alpha \neq fu,T} 0^{m_\alpha} - \sum_{n=1}^{N_Q} \prod_{\alpha=1}^{d} \phi_{\alpha,n}^{m_\alpha} W_\alpha \right] \qquad 1.8$$

Therefore the solution of the problem involves solving the ODEs (1.6) with the source terms for each time step given by solution of a matrix equation.

In terms of “statistical reduction” that we mentioned in the previous section, DQMOM is analogous to a Monte Carlo method with N_Q particles. For very small N_Q, this would seem to be a great reduction in computational cost. However at each time step, a matrix of size $N_Q(d+1)$ X $N_Q(d+1)$ needs to be inverted. Matrix inversion is a computationally expensive procedure and this method loses its efficiency if N_Q or d are very large. DQMOM only ensures that some of the moments of the pdf evolve consistently. If the moment equations involve unclosed terms, one might need a larger value for N_Q to accurately represent the unclosed terms. In combustion problems, the chemical source term involves an exponential dependence in temperature, which is one of the internal variables, and this term cannot be closed with a finite number of moments. In this case there is some error in the DQMOM solutions. Another issue is the rank deficiency of the matrix A. It turns out that for a given choice of N_Q and d, it is not possible to evolve some sets of $N_Q(d+1)$ moments due to the singularity of the matrix

(Fox (2003)). The simplest example is for the case where $N_Q = d = 2$, in which it is not possible to evolve the most natural choice of moments M_{m_1,m_2} with (m_1,m_2)= (0,0),(1,0),(0,1),(1,1),(2,0),(0,2). In the formulation above, we made no mention of which moments to choose. The choice of moments can be arbitrary and further there may not be an optimal choice of moments (Upadhyay and Ezekoye (2005)). Hence to impose some order in the choice of moments and at the same time to discard the moments that give rise to a singular matrix, we propose the method of Selective Graded Lexicographic Ordering (SGLO).

The graded lexicographic order is an ordering scheme for multivariate polynomials. In our context we want to order multivariate moments of the form $M_{m_1,m_2,\ldots,m_d}$. The definition of graded lexicographic order (glex) is as follows (e.g. Dunkl and Xu (2001)). Let $m = (m_1, m_2, \ldots, m_d)$ and $m^* = (m_1^*, m_2^*, \ldots, m_d^*)$ be two d-tuples with $|m| = \sum_{i=1}^{d} m_i$ then $M_m \succ_{glex} M_{m^*}$ if $|m| > |m^*|$ or if $|m| = |m^*|$ then the first non zero entry in the difference $m - m^*$ is positive. This type of ordering ensures that the 0th order moment is tracked before all the 1st order moments. And all 1st order moments are tracked before the 2nd order moments (which provide information on pair correlations) and so on. However a strict ordering using this scheme is not possible because some moments cause the resulting matrix to be singular. Hence a selective ordering is necessary. In practice a set of d-tuples, ordered according to glex, is generated; then, as the matrix A is built up row by row, a singular value decomposition of the matrix is performed. If the condition number of the matrix becomes very large then the current row associated with a particular d-tuple is discarded and the next d-tuple in the order is chosen. This procedure, which we have called Selective Graded Lexicographic Ordering (SGLO), ensures that the matrix A in (1.7) is non-singular. As an example let us take the case $N_Q = 2, d = 3$ which is the case of the 2-step mechanism with two quadrature points. The set of eight valid moments is given by M_{m_1,m_2,m_3} with $(m_1, m_2, m_3) =$ (0,0,0), (1,0,0),(0,1,0),(0,0,1),(2,0,0),(1,1,0),(3,0,0). Note that it is not possible to track a few of the second order moments such as (0,1,1), (1,0,1), (0,0,2) etc. If some of the moments carry essential information for a particular application then they can be selected and the remaining moments can be ordered using SGLO. The ordering we have discussed is not the only possible ordering. The optimal choice of moments may depend on the problem and may require trial and error to discover.

2. Chemical reaction models.

To investigate the performance of DQMOM for low and high dimensional systems we consider chemical reaction mechanisms of increasing complexity. The chemical reaction model is required to evaluate the chemical source terms $S_i(\vec{\psi})$ appearing in equation 1.6.

2.1. One step chemistry mechanism.

This is the simplest possible mechanism. We use the global 1-step reaction for propane combustion from Westbrook and Dryer (1981).

$$C_3H_8 + 5O_2 + 18.8N_2 \rightarrow 3CO_2 + 4H_20 + 18.8N_2 \qquad 2.1$$

In this case d=2 (bivariate problem). One can solve for $\phi_1 = \phi_{C_3H_8}$ and $\phi_2 = \phi_{CO_2}$. The mass fraction of other species and temperature are linear functions of these two variables. The reaction rate is given by $R = W_F A[F]^a [O]^b \exp\left(\frac{-E_a}{RT}\right)$ with $[F] = \left(\frac{\rho\phi_F}{W_F}\right)$ etc, $A = 8.6\times10^{11}$, $E_a = 30.0 kcal/mol$, $a = 0.1$ and $b = 1.65$. The chemical source terms for ϕ_1 and ϕ_2 are $S_1 = \frac{-R}{\langle\rho\rangle}$ and $S_2 = \left(\frac{W_{CO_2}}{W_F}\right)\frac{R}{\langle\rho\rangle}$.

2.2. Two step chemical mechanism.

We use the 2-step mechanism for propane taken from Westbrook and Dryer (1981).

$$\begin{aligned} &C_3H_8 + 3.5O_2 + 13.16N_2 \rightarrow 3CO + 4H_20 + 13.16N_2 \\ &CO + 0.5O_2 \rightarrow CO_2 \end{aligned} \qquad 2.1$$

In this case $d = 3$, i.e. only three variables are sufficient for describing the chemical system. For instance one can choose $\phi_1 = \phi_{C_3H_8}, \phi_2 = CO, \phi_3 = CO_2$, then the mass fractions of the remaining species and temperature can be obtained from these variables by linear transformations. The reaction rates for the two reaction steps are given by

Step 1. $R_1 = W_F A_1 [F]^a [O]^b \exp\left(\frac{-E_{a1}}{RT}\right)$

Step 2. $R_2 = W_{CO}\left\{A_{2f}[CO]^c [H_2O]^d [O_2]^e - A_{2b}[CO_2]^f\right\}\exp\left(\frac{-E_{a2}}{RT}\right)$

Where $[F] = \left(\frac{\rho\phi_F}{W_F}\right)$ denotes the molar concentration of fuel etc. $a = 0.1, b = 1.65, c = 1.0, d = 0.5, e = 0.25, f = 1.0$. Activation energies for the various reactions are $E_{a1} = 30.0 kcal/mol, E_{a2} = 40.0 kcal/mol$. Pre-exponential factors are $A_1 = 1.0\times10^{12}, A_{2f} = 10^{14.6}, A_{2b} = 5\times10^8$.

Then the chemical source terms for the variables ϕ_1, ϕ_2 and ϕ_3 are $S_1 = \frac{-R_1}{\langle\rho\rangle}$, $S_2 = \frac{R_1 - R_2}{\langle\rho\rangle}$ and $S_3 = \left(\frac{W_{CO_2}}{W_{CO}}\right)\frac{R_2}{\langle\rho\rangle}$.

2.3. More Detailed Reaction mechanisms.

We also consider two more detailed mechanisms for the combustion of methane. The first is J-Y Chen's reduced 12 step mechanism (from the GRI 2.11 mechanism). This mechanism provides reaction rates for 16 species. The dimension is 16 when one includes the temperature and with the constraint of the mass fractions of species summing to one. The second is the ARM2 mechanism which is also based on GRI 2.11. This mechanism provides reaction rates for 19 species, so that d=19. These mechanisms can be found in the TNF website maintained by Sandia (http://www.ca.sandia.gov/TNF/chemistry.html).

3. Computation and Results.

We present the comparison of solutions obtained by Monte Carlo simulations and DQMOM for the Partially Stirred Reactor Model. For the DQMOM computations, equations (1.6) are integrated using an explicit scheme with step size $\Delta t = 0.1 \times \min(\tau_{res}, \tau_{mix})$. The chemical source term is integrated using a much smaller time step. For the 1-step and 2-step mechanisms, a higher order adaptive Runge-Kutta method (Press et al. (1992)) is used. For the 12 step and ARM2 mechanism, an explicit scheme is used with Δt as small as 10^{-7}. The Monte Carlo simulations are performed using the method given in Ren and Pope (2004) with the exception that ISAT is not used for evaluating the chemical source term.

Figure 1 shows the plot of the mean and variance of the mixture fraction with time. The mixture fraction does not depend on the reaction scheme used but describes the mixing of the inlet streams of fuel and oxidizer and the air initially present in the reactor. For the pure mixing problem, there are no errors due to moment closure and therefore the means and variances predicted by DQMOM is also the exact solution. Also shown is the Monte Carlo Solution which shows some fluctuations about even for 100,000 particles. In figure 2 we plot the means and variances of carbon dioxide (the product) computed using DQMOM with two quadrature points and Monte Carlo simulations. In this case there is a slight difference in the means in the final stationary state as well as during the evolution. There is a larger error in prediction of the variance with the variance predicted using Monte Carlo simulations being almost five times larger. This indicates that although the mean is predicted accurately, there can be significant error in the variance.

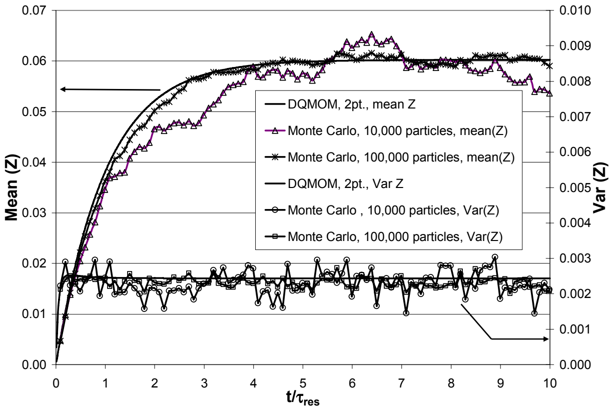

Figure 1. Mean and variance of mixture fraction computed using 2pt. DQMOM and Monte Carlo simulations for the 1step mechanism.

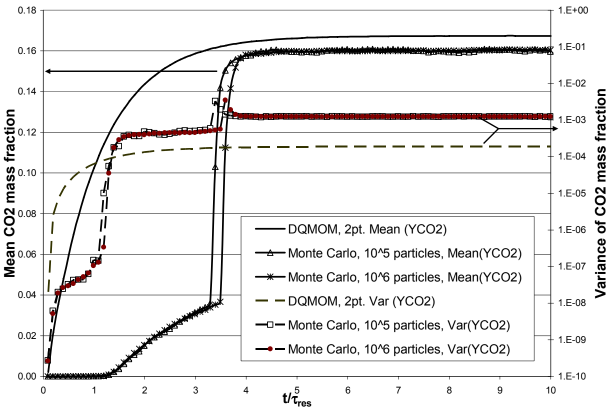

Figure 2. Mean and variance of carbon dioxide mass fraction computed using 2pt. DQMOM and Monte Carlo simulations for the 1-step mechanism

Figure 3 shows the mean mass fractions of the different species for the two step reaction mechanism. The profiles are computed using 2 point DQMOM. In figure 4 we plot the variances of the species mass fractions. Both figure 1 and 2 correspond to the case of equivalence ratio $\Phi = 1$ and $\tau_{res} = 1; \tau_{mix} = 0.1$. Initially the carbon monoxide concentration peaks due to the first reaction and as the temperature increases the second reaction converts CO into CO2.

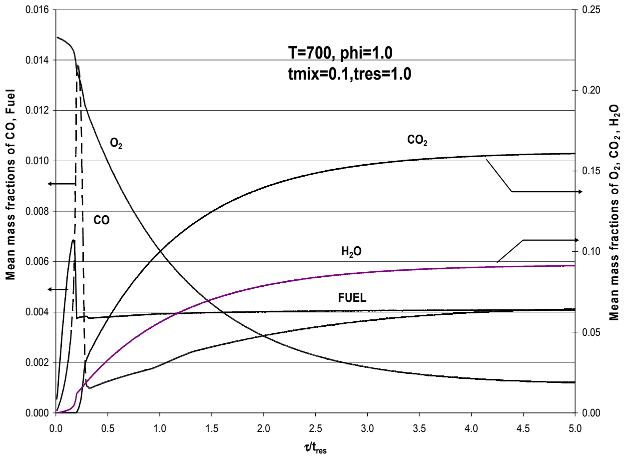

Figure 3. Mean of species mass fractions computed using 2 point DQMOM for the 2 step reaction mechanism.

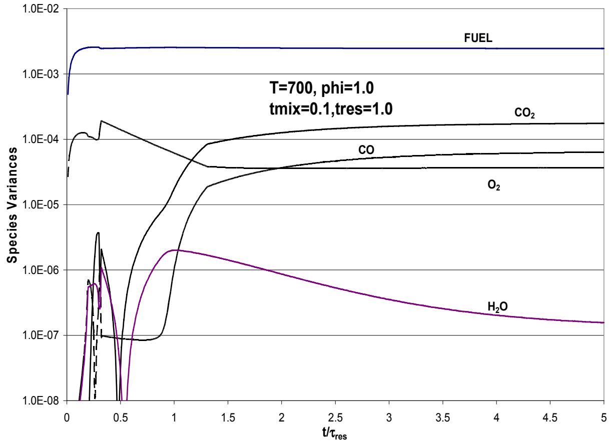

Figure 4. Variance of species mass fractions computed using 2 point DQMOM for the 2 step reaction mechanism.

Figure 5 shows the mean species profiles for the ARM2 mechanism computed using 2-point DQMOM. The simulations are run for a constant temperature of 1500K, equivalence ratio $\Phi = 1$ and $\tau_{res} = 1; \tau_{mix} = 0.1$. This is a very high dimensional case but the computations indicate that it may be possible to apply DQMOM without getting into problems due to the ill-conditioning of the matrices. Comparison of these results with Monte Carlo simulations remains a future task.

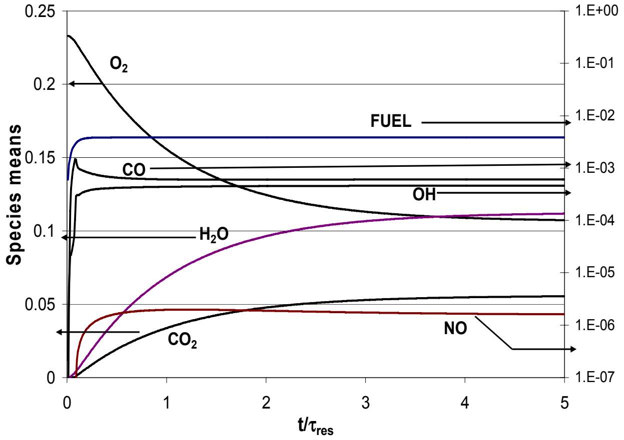

Figure 5. The means of some major and trace species computed using the 2-point DQMOM method for the ARM2 mechanism. The temperature is kept fixed at 1500K.

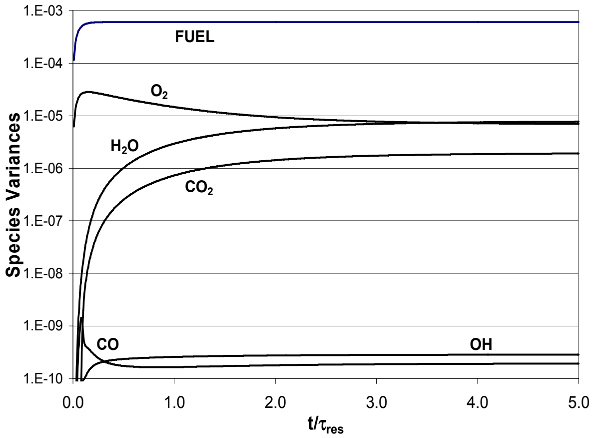

Figure 6. Variances of the major species and some significant trace species computed using 2-point DQMOM method for the ARM2 mechanism. The temperature is kept fixed at 1500K.

4. Conclusions.

In this work we have implemented the Direct Quadrature Method of Moments for turbulent mixing and combustion problems. We find that DQMOM can be used for multivariate problems of high dimension provided that moments are chosen such that the matrices involved in the computation are not ill-conditioned or singular. Stochastic simulations in spaces of high dimension are important in a number of fields of chemical engineering. These simulations can be very demanding in terms of computational cost. The use of moment methods such as DQMOM can provide a simplification in the computations. In combustion simulations, there is a great reduction in the number of times the chemical source term needs to be evaluated. For DQMOM, the chemical source term needs to be evaluated only for the quadrature points or “environments”. Also a minimum of two or three quadrature points can be used for each dimension. As the ODEs describing chemical reactions are very stiff, the evaluation of the chemical source term constitutes the greatest computational cost for a turbulent reactive flow simulation using Monte Carlo methods. Hence in this aspect DQMOM offers some advantages. Of course, high-dimensional problems require significant computational cost even when simplified moment methods are used. For problems with high dimensions, such as the 19 dimensional problem considered in this study, the number of moments that need to be tracked goes up. In a CFD simulation of turbulent flow, one has to solve a larger number of partial differential equations. There is also the cost of inverting a matrix of size $N_Q(d+1)$ which becomes prohibitively expensive as N_Q or d goes up. Use of efficient but approximate methods of matrix inversion along with parallelization of the code can further speed up the process. In the future, the comparison of solutions obtained using DQMOM with Monte Carlo simulations, determination of optimal choices of moments and evaluation of the error incurred in moment closure schemes needs to be carried out.

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