Dimension Reduction and Tabulation of Combustion Chemistry using ICE-PIC and ISAT

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Abstract— **Progress is reported in the integration of two methodologies to enable the efficiency application of realistic combustion chemistry in computational fluid dynamics. These methodologies are ICE-PIC (invariant constrained-equilibrium edge manifold using the pre-image curve method) for dimension reduction, and ISAT (*in situ* adaptive tabulation) for tabulation of the reduced system. New results are reported on the tangent vectors of the constrained-equilibrium and ICE manifolds, which are important quantities in ICE-PIC/ISAT. The test case of a partially-stirred reactor with methane combustion is used to demonstrate the accuracy and efficiency of the combined approach.**

I. INTRODUCTION

Dimension reduction is essential to the use of detailed chemical kinetics in computations of combustion and many other reactive flows. Modern chemical mechanisms for hydrocarbon fuel may contain of order 1,000 species [1], and it is clearly impracticable to use this detailed information directly in multi-dimensional computational fluid dynamics (CFD) calculations. A combination of three approaches that enables the use of detailed chemical information consists of: (1) reduction to a skeletal mechanism [2], [3], [4] involving of order 100 species; (2) dimension reduction (DR) to reduce the number of degrees of freedom to of order ten; and (3) tabulation to significantly reduce the cost of expensive evaluations, e.g., the integration of ordinary differential equations (ODEs). In this work we consider the integration of two successful techniques, namely, the invariant constrained-equilibrium edge pre-image curve (ICE-PIC) method for dimension reduction [5], [6], [7], and *in situ* adaptive tabulation (ISAT) [8], [9].

In the next section we briefly review the ICE-PIC method as implemented in conjunction with ISAT. Then we derive expressions for the tangent vectors of the constrained-equilibrium manifold (CEM) and the ICE manifold, which are needed by ISAT. Finally, at the Workshop results will be given for the test case of a partially-stirred reactor, showing the accuracy of the dimension reduction and the efficiency gains achieved by ISAT.

II. THE ICE-PIC METHOD

We give here a succinct overview of the ICE-PIC method, as it is implemented in conjunction with ISAT. More details can be found in [5], [6], [7].

We consider a gas-phase mixture of n_s chemical species composed of n_e elements. The thermochemical state of the mixture (at a given position and time) is completely characterized by the pressure p, the species enthalpy h, and the n_s-vector $\mathbf{z}$ of the specific moles of the species. To simplify the exposition, we take p and h to be given constants, and so the state is given by $\mathbf{z}$.

Due to chemical reactions, the composition evolves by

$$\frac{d\mathbf{z}}{dt} = \mathbf{S}(\mathbf{z}), \tag{1}$$

where $\mathbf{S}$ is the n_s-vector of chemical production rates. The "reaction mapping" $\mathbf{R}(\mathbf{z}, t)$ is defined to be the solution to (1) after time t from the initial condition $\mathbf{z}$. And the mapping gradient $\mathbf{A}(\mathbf{z}, t)$ is the $n_s \times n_s$ matrix with components

$$A_{ij} = \partial R_i / \partial z_j. \tag{2}$$

In practice $\mathbf{R}$ and $\mathbf{A}$ are obtained together using the ODE solver DASAC [10].

In the ICE-PIC method, the species are decomposed as $\mathbf{z} = \{\mathbf{z}^r, \mathbf{z}^u\}$, where $\mathbf{z}^r$ is an n_{rs} vector of "represented" species, and $\mathbf{z}^u$ is an n_{us}-vector of "unrepresented" species (with $n_{rs} + n_{us} = n_s$ and $n_{rs} < n_s - n_e$). At the present stage of development of the methodology, the represented species are specified: ultimately, the methodology should identify the optimal specification. The "reduced representation" of the species used in ICE-PIC is $\mathbf{r} \equiv \{\mathbf{z}^r, \mathbf{z}^{u,e}\}$, where $\mathbf{z}^{u,e}$, is an n_e-vector giving the specific moles of the elements in the unrepresented species. Thus $\mathbf{r}$ is a vector of length $n_r = n_{rs} + n_e$, and the dimensions of the system is reduced from n_s to $n_r < n_s$. This dimension reduction process can be written

$$\mathbf{r} = \mathbf{B}^T \mathbf{z}, \tag{3}$$

where $\mathbf{B}$ is a known constant $n_s \times n_r$ matrix.

The fundamental issue in dimension reduction of combustion chemistry is "species reconstruction" that is, given $\mathbf{r}$, define an appropriate full composition $\mathbf{z}$. We denote by $\mathbf{z}^{ICE}(\mathbf{r})$ the species reconstruction given by the ICE-PIC method. We also consider $\mathbf{z}^{CE}(\mathbf{r})$ which is the constrained-equilibrium (maximum-entropy) composition, as used in the rate-controlled constrained equilibrium method (RCCE, [11], [12], [13]). This is readily computed using the constrained-equilibrium code CEQ [14].

In the n_r-dimensional reduced space, the "realizable region" is the convex polytope in which each component of $\mathbf{r}$ is non-negative. Its boundary consists of at most n_r facets on which one component of $\mathbf{r}$ is zero. The "constrained equilibrium edge" is defined as $\mathbf{z}^{CE}(\mathbf{r})$ for all $\mathbf{r}$ on the boundary. The ICE manifold is defined as $\mathbf{R}(\mathbf{z}^{CE}(\mathbf{r}), t)$ for all $\mathbf{r}$ on the boundary and all $t \geq 0$. Thus the ICE manifold is the trajectory-generated manifold originating from all the constrained equilibrium compositions on the boundary. Some important properties of the ICE manifold are:

1) existence: for all realizable $\mathbf{r}$ there exists a manifold point $\mathbf{z}^{ICE}(\mathbf{r})$
2) invariance: the ICE manifold is invariant with respect to (1)
3) continuity: the ICE manifold is continuous
4) smoothness: the ICE manifold is piecewise smooth, and is the union of smooth manifolds generated by the facets
5) uniqueness: for a reasonable specification of the represented species, the manifold is not "folded", so that for given $\mathbf{r}$ there is a unique manifold point $\mathbf{z}^{ICE}(\mathbf{r})$.

Provided that the manifold is not folded, given a realizable value of $\mathbf{r}$, there is a unique "generating boundary point" $\mathbf{r}^g$, and time τ such that

$$\mathbf{z}^{ICE}(\mathbf{r}) = \mathbf{R}(\mathbf{z}^{CE}(\mathbf{r}^g), \tau). \tag{4}$$

The pre-image curve method is used to identify $\mathbf{r}^g$ (given $\mathbf{r}$). Of course, consistency conditions are

$$\mathbf{B}^T \mathbf{z}^{ICE}(\mathbf{r}) = \mathbf{B}^T \mathbf{z}^{CE}(\mathbf{r}) = \mathbf{r}. \tag{5}$$

At the Workshop, the presentation will focus on an exposition of the ICE-PIC/ISAT methodology and on its performance for the test case described in Sec.V. In the next two sections, we present some new theoretical results which provide quite simple means of determining the tangent vectors of the constrained-equilibrium and ICE manifolds.

III. The CEM Tangent Vectors

An important quantity in the ICE-PIC method is the $n_s \times n_r$ matrix $\mathbf{T}^{CE}$ whose columns span the tangent space of the CE manifold, and which relates infinitesimal changes in $\mathbf{z}^{CE}$ to those in $\mathbf{r}$ by

$$d\mathbf{z}^{CE} = \mathbf{T}^{CE} d\mathbf{r}. \tag{6}$$

We have obtained a new, simple expression for $\mathbf{T}^{CE}$. It is presented here for the case of fixed pressure and temperature, from which the corresponding result for fixed p and h is readily obtained.

For the case considered, the constrained equilibrium composition is given by [14]

$$\mathbf{z}^{CE} = \bar{N} \exp\left(-\tilde{\mathbf{g}} + \mathbf{B}\boldsymbol{\lambda}\right), \tag{7}$$

where $\bar{N} = \sum_{i=1}^{n_s} z_i^{CE}$ are the specific moles of all species; $\tilde{\mathbf{g}}$ are normalized Gibbs functions; and $\boldsymbol{\lambda}$ are constraint potentials (or Lagrange multipliers).

Considering infinitesimals, we obtain from (7)

$$d\mathbf{z}^{CE} = \mathbf{z}^{CE} d\, \ln(\bar{N}) + \mathbf{Z}\mathbf{B}d\boldsymbol{\lambda}, \tag{8}$$

where $\mathbf{Z}$ is the diagonal matrix formed from $\mathbf{z}^{CE}$. Summing (8) over all the species leads to the constraint

$$0 = \mathbf{z}^T \mathbf{B} d\boldsymbol{\lambda} = \mathbf{r}^T d\boldsymbol{\lambda}. \tag{9}$$

Equation (8) can be re-expressed as

$$d\mathbf{z}^{CE} = \mathbf{M} d\hat{\boldsymbol{\lambda}}, \tag{10}$$

with

$$d\hat{\boldsymbol{\lambda}} \equiv d\boldsymbol{\lambda} + \frac{\mathbf{r} d\bar{N}}{|\mathbf{r}|^2 \bar{N}}, \tag{11}$$

and

$$\mathbf{M} \equiv \mathbf{z}\mathbf{r}^T + \mathbf{Z}\mathbf{B}\left(\mathbf{I} - \frac{\mathbf{r}\mathbf{r}^T}{|\mathbf{r}|^2}\right). \tag{12}$$

We observe from (10) that the columns of $\mathbf{M}$ span the tangent space. Let $\mathbf{W}$ denote any $n_s \times n_r$ matrix with $span(\mathbf{W}) = span(\mathbf{M}) = span(\mathbf{T}^{CE})$. Then there exists a non-singular $n_r \times n_r$ matrix $\mathbf{D}$ such that $\mathbf{T}^{CE} = \mathbf{W}\mathbf{D}$. From (5) we obtain

$$\mathbf{B}^T d\mathbf{z}^{CE} = d\mathbf{r} = \mathbf{B}^T \mathbf{T}^{CE} d\mathbf{r} = \mathbf{B}^T \mathbf{W}\mathbf{D} d\mathbf{r}, \tag{13}$$

and hence

$$\mathbf{B}^T \mathbf{T}^{CE} = \mathbf{I}, \tag{14}$$

$$\mathbf{D} = (\mathbf{B}^T \mathbf{W})^{-1}, \tag{15}$$

and finally

$$\mathbf{T}^{CE} = \mathbf{W}(\mathbf{B}^T \mathbf{W})^{-1}. \tag{16}$$

In practice $\mathbf{W}$ is best taken as an orthonormal basis for $span(\mathbf{T}^{CE})$, obtained from the SVD or QR decomposition of $\mathbf{M}$.

It is interesting to observe that $\mathbf{T}^{CE}$ is solely determined by $\mathbf{z}^{CE}$ and $\mathbf{B}$, and does not otherwise depend on any thermodynamic information (such as p, T or $\tilde{\mathbf{g}}$).

IV. The ICE Manifold Tangent Vectors

Also important in combining ISAT with ICE-PIC is the matrix of ICE manifold tangent vectors $\mathbf{T}^{ICE}$ defined such that

$$d\mathbf{z}^{ICE} = \mathbf{T}^{ICE} d\mathbf{r}. \tag{17}$$

We are considering now the relevant case of constant pressure and enthalpy, so that (6) and (17) are at fixed p and h. (This implies a re-definition of $\mathbf{T}^{CE}$.)

From (4), considering infinitesimal changes $d\mathbf{r}^g$ and $d\tau$, we have correspondingly

$$d\mathbf{z}^{CE} = \mathbf{T}^{CE} d\mathbf{r}^g, \tag{18}$$

and

$$d\mathbf{z}^{ICE} = \mathbf{A}(\mathbf{z}^g, \tau)\mathbf{T}^{CE}(\mathbf{r}^g) d\mathbf{r}^g + \mathbf{S}(\mathbf{z}^{ICE}) d\tau. \tag{19}$$

Let k denote the index of the component of $\mathbf{r}^g$ which is zero on the boundary facet, i.e. $r_k^g = 0$. Since we require $\mathbf{r}^g + d\mathbf{r}^g$ to be on the boundary, it follows that dr_k^g is zero. This consideration and (19) show that the tangent space of the ICE manifold is spanned by $\mathbf{S}(\mathbf{z}^{ICE})$ and the $n_r - 1$ vectors obtained from $\mathbf{AT}^{CE}$, with the k-th column omitted. Then, by the same argument that leads to (16), we have

$$\mathbf{T}^{ICE} = \hat{\mathbf{W}}(\mathbf{B}^T \hat{\mathbf{W}})^{-1}, \tag{20}$$

where $\hat{\mathbf{W}}$ is an $n_s \times n_r$ matrix (obtained from $\mathbf{S}$ and $\mathbf{AT}^{CE}$) which spans the ICE manifold tangent space.

V. Results

At the Workshop, results will be presented for the test case of a partially-stirred reactor (PaSR) with methane combustion [9]. The results quantify the dimension reduction errors in the ICE-PIC and RCCE as functions of the number of represented variable, n_r. Also, the efficiency of the ISAT implementation is characterized in terms of table size and retrieve time.

VI. Conclusions

The combination of ICE-PIC and ISAT offers accurate dimension reduction and efficient tabulation. Advances have been made both in the theory (e.g., in the accurate and efficient evaluation of the tangent vectors) and in the computational implementation.

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