Diffusion Effects on Slow Invariant Manifolds

Joshua D. Mengers*, Joseph M. Powers
Department of Aerospace and Mechanical Engineering
University of Notre Dame
Notre Dame, IN 46556

Abstract

Diffusion effects on the Slow Invariant Manifold (SIM) of a closed reactive system are examined with the goal of developing a model reduction technique which rigorously accounts for the coupling of reaction and diffusion processes. A robust method of constructing a one-dimensional SIM by calculating equilibria and then integrating heteroclinic orbits is extended to systems with diffusion across a small characteristic length. First, a spatially homogeneous system of NO production is discussed. Diffusion is then added as a correction to the spatially homogeneous system using a Galerkin method to project the infinite dimensional dynamical system onto a low dimensional approximate inertial manifold. A critical length is identified, above which a perturbed SIM is found by similar techniques of calculating equilibria and integrating heteroclinic orbits.

Introduction

Reactive flow problems are known to display multiscale phenomena that cause challenges in the numerical simulations of such problems. Verification of these simulations requires grid resolution that captures the full range of scales in both space and time. Large disparity in scales induces simulations that require significant computational effort. A disparity in temporal scales can be caused by the reaction mechanism alone, while the addition of diffusion couples the differing reaction time scales to a disparity in length scales. Recently, considerable effort has been expended in identification of model reduction techniques for reactive flows in order to reduce the computational cost, while maintaining as much consistency with the underlying reactive flow physics as possible.

The reviews of Griffiths [1] or Lu and Law [2] are good references for these techniques in general. Most of the methods described therein address only reaction mechanisms. Some current research that extends these methods to systems with diffusion are Singh, et al. [3], Ren and Pope [4], Davis [5, 6], Bykov and Maas [7], Lam [8], Adrover, et al. [9], and Goussis, et al. [10]

The study of Davis and Skodje [11] is particularly relevant. In their study, which was performed on spatially homogeneous reactive systems, the authors calculate a one-dimensional Slow Invariant Manifold (SIM) of the system by integrating a heteroclinic orbit between the system's physical and non-physical equilibrium points. This technique has recently been refined by Al-Khateeb et al. [12] to examine larger systems. The SIM is a unique trajectory of the dynamical system that describes the long time dynamics of the system's evolution efficiently, which makes the SIM one of the premiere reduction techniques available; however, the SIM has almost exclusively been used for spatially homogenous systems.

Specific Objectives

The objective of this paper is to examine how the addition of diffusion to a reactive system affects the SIM. Here, we focus on short length scales; in the limit of an infinitesimal length, diffusion will have a negligible effect, and the system will remain spatially homogeneous. We aim to identify a critical length scale at which diffusion has a significant effect on the spatially homogeneous system. We find that at this critical length a bifurcation occurs in the SIM. This critical length can serve as a rough division between what Goussis et al. [10] refer to as 'local' or 'global' analysis. The objective of the current paper will be to focus on the global analysis, where the time scales from diffusion modification are faster than the reaction time scales.

Methodology

By the addition of a linear Fick's law diffusion model to a spatially homogeneous reaction mechanism, the governing equations change from ordinary differential equations (ODEs),

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

to partial differential equations (PDEs),

$$\frac{\partial \mathbf{z}}{\partial t} = \mathbf{f}(\mathbf{z}) + \mathcal{D}\frac{\partial^2 \mathbf{z}}{\partial x^2}. \tag{2}$$

*Corresponding author: jmengers@nd.edu
Proceedings of the 2010 Technical Meeting of the Central States Section of the Combustion Institute

Reaction	a	β	E
$N + NO \leftrightharpoons N_2 + O$	2.107×10^{13}	0.00	0.0
$N + O_2 \leftrightharpoons NO + O$	5.839×10^{9}	1.01	6196

Table 1: The Zel'dovich mechanism for NO production. The units for a are $mol/(cm\ s\ K)$, E are cal/mol, and β is dimensionless.

A_1	6.4×10^{-2}	B_1	-4.2×10^{4}	C_1	9.1×10^{6}
D_1	-3.1×10^{12}	E_1	5.5×10^{11}		
A_2	6.4×10^{-2}	B_2	3.8×10^{4}	C_2	-9.1×10^{6}
D_2	-2.0×10^{12}	E_2	-5.5×10^{11}		

Table 2: Coefficients for Eqs. (6-8); the units of A_i are $mol/(g\ s)$, B_i and C_i are $1/s$, D_i and E_i are $g/(mol\ s)$.

The term $\mathbf{z} \in \mathbb{R}^R$ represents a vector of length R, whose ith term gives the number of moles per unit mixture mass of species i, and the function $\mathbf{f} \in \mathbb{R}^R$ represents a vector of length R, whose ith term gives the reaction source term for species i, and $\mathcal{D}$ is the assumed constant diffusion coefficient. For our analysis the boundary conditions are chosen as $\partial\mathbf{z}/\partial x|_{x=0} = \partial\mathbf{z}/\partial x|_{x=L} = 0$. Attention is restricted to a domain $x \in [0, L]$, $t \in [0, \infty)$, where we choose L as a small length parameter.

We apply a Galerkin projection to this system, projecting it onto an approximate inertial manifold [13]. To accomplish this we assume a spectral decomposition of $\mathbf{z}$

$$\mathbf{z} = z_i = \sum_{m=0}^{\infty} z_{i,m}(t)\phi_m(x), \quad i = 1, \ldots, R, \quad (3)$$

where $z_{i,m}(t)$ is the mth time-dependent amplitude associated with species i, and $\phi_m(x)$ are the corresponding basis functions. The ideal choice for ϕ_m are the eigenfunctions of the diffusion operator that match the boundary conditions; for our choice of boundary conditions, the complete basis is

$$\phi_n = \cos\left(\frac{n\pi}{L}x\right), \quad n = 0, 1, \ldots, \infty, \quad (4)$$

which has corresponding eigenvalues $\mu_n = (n\pi/L)^2$. Because the Fick's law diffusion operator is self-adjoint, our basis functions are orthogonal, and their eigenvalues are real. We then substitute Eq. (3) into Eq. (2), and take the inner product with each basis function, ϕ_n, yielding the infinite system of ODEs

$$\frac{dz_{i,n}}{dt} = \frac{<\phi_n, \mathbf{f}(\sum_{m=0}^{\infty} z_{i,m}\phi_m)>}{<\phi_n, \phi_n>} - \mu_n \mathcal{D} z_{i,n}. \quad (5)$$

When we truncate this system at a specific $n = N$, this method projects the infinite dimensional PDE onto a finite dimensional approximate inertial manifold. On this approximate inertial manifold, the standard technique of connecting heteroclinic orbits to find the SIM [11, 12] is applied. Importantly, the heteroclinic orbit originates along the unstable eigenvector of a saddle point equilibrium that has one positive eigenvalue and connects to the physical equilibrium sink along its slowest eigenvector.

For the present analysis, we choose to truncate this series at $N = 1$. To maintain full resolution of the spatio-temporal dynamics for this choice of N, we must restrict our domain length to sufficiently small L. Furthermore, the $N = 1$ case is adequate since a choice of a larger value of N does not inherently change the results and $N = 1$ allows concise illustration of our conclusions.

Zel'dovich Model

We evaluate the Zel'dovich mechanism of NO production similar to the system studied in [12]; it is shown in Table 1. This system is taken to be isothermal and isochoric, at $T = 3500\ K$ and $V = 10^3\ cm^3$. Each of the five species are initialized with $10^{-3}\ mol$ uniformly in space. The constraints in the mechanism make this system isobaric and allow the system to be reduced to only two variables; we choose to evaluate the specific moles of NO and N as z_1 and z_2, respectively. The governing equations for this system are two PDEs

$$\frac{\partial z_i}{\partial t} = A_i + B_i z_1 + C_i z_2 + D_i z_1 z_2 + E_i z_2^2 + \mathcal{D}\frac{\partial^2 z_i}{\partial x^2}, \quad i = 1, 2, \quad (6)$$

where the coefficients are given in Table 2; the diffusion coefficient, $\mathcal{D} = 1.87 \times 10^1\ cm^2/s$, is chosen as an *ad hoc* average of species Fick's law diffusion coefficients from the CHEMKIN TRANSPORT database [14].

Applying the Galerkin projection to this model and truncating at $N = 1$ modifies the governing equations to a system of four ODEs

$$\begin{aligned} \frac{dz_{i,0}}{dt} &= A_i + B_i z_{1,0} + C_i z_{2,0} + D_i z_{1,0} z_{2,0} + \\ &\quad D_i \frac{z_{1,1} z_{2,1}}{2} + E_i z_{2,0}^2 + E_i \frac{z_{2,1}^2}{2}, \quad (7) \\ \frac{dz_{i,1}}{dt} &= B_i z_{1,1} + C_i z_{2,1} + D_i z_{1,0} z_{2,1} + \\ &\quad D_i z_{1,1} z_{2,0} + 2E_i z_{2,0} z_{2,1} - \frac{\pi^2}{L^2}\mathcal{D} z_{i,1}, \quad (8) \\ &\quad i = 1, 2. \end{aligned}$$

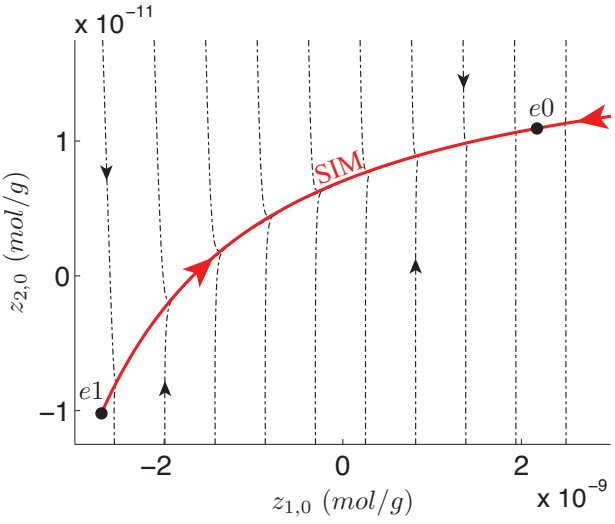

Figure 1: The spatially homogeneous SIM for the Zel'dovich mechanism.

In Eq. (7) the evolution $z_{i,0}$ term remains identical to the spatially homogeneous system when $z_{i,1} = 0$ for all i. This means that all the spatially homogenous equilibria remain in the diffusion-modified system.

Results and Discussion

In the limit as $L \to 0$, the $z_{i,1}$ terms decay to zero infinitely fast in time, reducing Eqs. (7,8) to the governing equations for a spatially homogeneous system. Each of this system's equilibria has two eigenvalues: λ_1^e and, λ_2^e. These equilibria are classified by their eigenvalues to construct the SIM. Further analysis will focus on two of this system's equilibria: the physical equilibrium, $e0$, which is a sink, and the equilibrium from which one branch of the SIM originates, $e1$, which is a saddle with one unstable mode. These two equilibria are shown in Fig. 1 along with the SIM and a set of random trajectories showing the SIM's attractiveness. The eigenvalues of these two equilibria are $\lambda_1^{e0} = -7.245 \times 10^4\ 1/s$, $\lambda_2^{e0} = -1.343 \times 10^7\ 1/s$, $\lambda_1^{e1} = 2.444 \times 10^5\ 1/s$, and $\lambda_2^{e1} = -3.977 \times 10^6\ 1/s$.

For small finite L, the system is described by Eqs. (7,8). The equilibria from the spatially homogeneous limit also are equilibria of this system; however, an analysis of their Jacobian will show they now have four eigenvalues, two identical to the spatially homogeneous eigenvalues, and two modified by diffusion: $\lambda_{1,0}^e = \lambda_1^e$, $\lambda_{2,0}^e = \lambda_2^e$, $\lambda_{1,1}^e = \lambda_1^e - \pi^2\mathcal{D}/L^2$, and $\lambda_{2,1}^e = \lambda_2^e - \pi^2\mathcal{D}/L^2$. From this analysis we find that $e0$ remains a sink, as all of its eigenvalues remain real and negative, and $e1$ remains a saddle; however, $e1$ has either one or two positive eigenvalues depending on the diffusion-modification term. Upon further investigation of the system, we find a bifurcation in the slow dynamics of the system at the critical length, L_c, where $e1$ switches between one and two positive eigenvalues and, therefore, has a zero eigenvalue. This occurs at

$$L_c = \pi\sqrt{\mathcal{D}/\lambda_1^{e1}}; \tag{9}$$

for our parameters we find $L_c = 2.745 \times 10^{-2}\ cm$.

To highlight the character of this bifurcation, let us compare two length scales: $L = L_1 = 1 \times 10^{-3}\ cm \ll L_c$, and $L = L_c$; the eigenvalues associated with these lengths are shown in Table 3. When $L = L_1$, $e1$ is a saddle with one positive eigenvalue, so it remains the critical point from which the SIM originates. When $L = L_c$, $e1$ is a non-hyperbolic critical point; at this length it is undergoing a transition from one to two positive eigenvalues. This transition is part of a bifurcation in which two additional equilibria appear for $L > L_c$. These additional equilibria each have only one positive eigenvalue, which have heteroclinic orbits that connect to the slow eigenvector of $e0$. This has ramifications on the SIM and the slow dynamics of the system. To illustrate this point the local time scales, $\tau = 1/\lambda$, are plotted in the neighborhood of

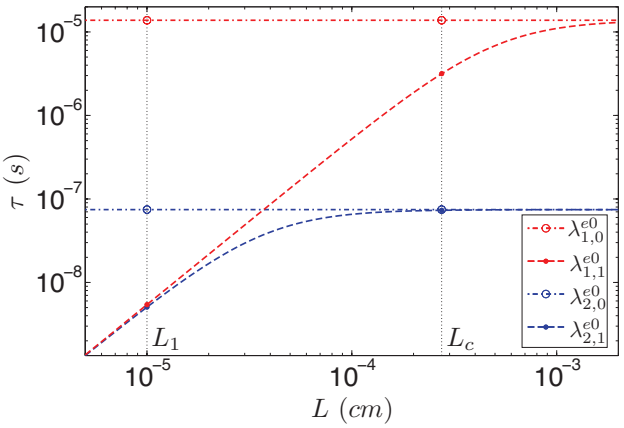

Figure 2: The time scales of the Galerkin projection of the Zel'dovich mechanism as a function of domain length.

	L_1		L_c	
	$e0$	$e1$	$e0$	$e1$
$\lambda_{1,0}$	-7.2×10^4	2.4×10^5	-7.2×10^4	2.4×10^5
$\lambda_{2,0}$	-1.3×10^7	-3.9×10^6	-1.3×10^7	-3.9×10^6
$\lambda_{1,1}$	-1.8×10^8	-1.8×10^8	-3.2×10^5	0.0
$\lambda_{2,1}$	-2.0×10^8	-1.9×10^8	-1.4×10^7	-4.2×10^6

Table 3: Eigenvalues of the two equilibria, $e0$ and $e1$, for the two length scales; the unit of λ are $1/s$.

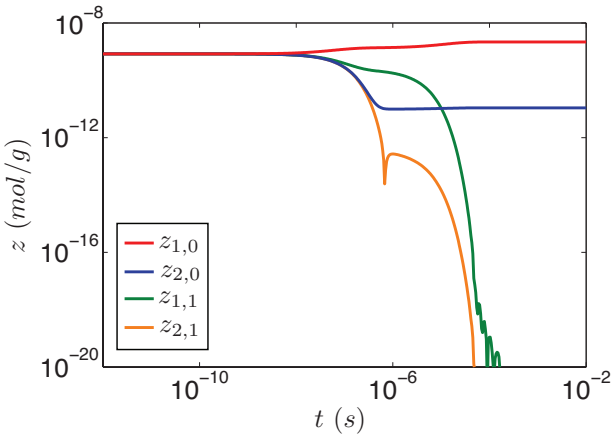

Figure 3: Evolution of Galerkin Coefficients.

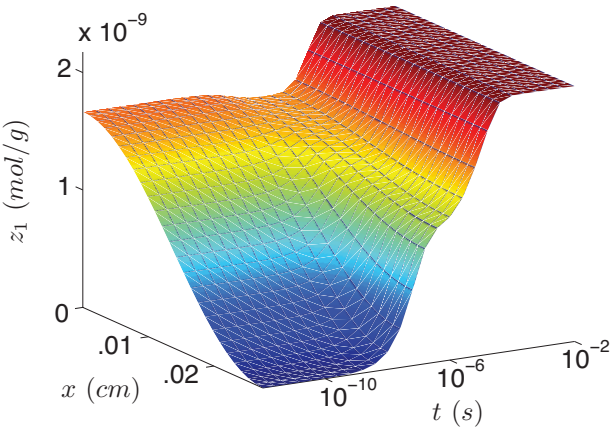

Figure 4: Evolution of the $N = 1$ Galerkin projection in space and time for $L = L_c$.

$e0$ in Fig. 2. Notice that for $L \ll L_c$, the diffusion-modification induces much faster time scales than either reaction only time scale. This implies that the slow dynamics are governed by the reaction only processes. When $L \approx L_c$ this is no longer the case, and the time scales from diffusion are infringing upon the time scales from reaction.

The evolution of the Galerkin coefficients for the $L = L_c$ case with the initial condition $z_{i,m} = 1.5 \times 10^{-9}\ mol/g$ is shown in Fig. 3. The decay in $z_{i,1}$ that occurs approximately the same time as the $z_{i,0}$ values are equilibrating shows that diffusion and reaction time scales are fully coupled for this length. The evolution of this $N = 1$ Galerkin method when reconstructed in x-t space is shown in Fig. 4

The dependency of diffusion-coupling on the considered domain length becomes even more evident when we look at a phase plane analysis of random trajectories for each of the lengths considered, shown in Fig. 5. In Figs. 5(a,c) each length is shown in the spatially homogeneous projection. The spatially homogenous SIM (red line) remains unchanged, still attracting the trajectories for each length. This implies that the slowest dynamics of the system remain unchanged.

The effects from the bifurcation become apparent in Figs. 5(b,d). In Fig. 5(b) the much faster diffusion time scales equilibrate quickly so the trajectories proceed to the $z_{1,0}$-$z_{2,0}$ plane, which is a two-dimensional SIM for this system. Once on that plane, the system follows the dynamics of the spatially homogeneous system toward, and then along the spatially homogeneous SIM to the equilibrium. In Fig. 5(d) the slower diffusion time scale prevents a similar relaxation, now causing the trajectories to relax to a surface which follows a curve similar to the spatially homogeneous SIM and projected in $z_{1,1}$ direction. This surface represents a two-dimensional SIM, which remains to be precisely calculated.

Conclusions

For small characteristic lengths, the effects of diffusion on a reactive system are minimal, allowing for a global analysis of the system, but when lengths are close to or above a critical length defined by this method, diffusion plays a major role. For lengths longer than the critical length, a diffusion-modified SIM governs the long time dynamics of these systems; which would suggest that the use of a local analysis would be advantageous. To maintain an accurate reduction technique, a global spatially homogeneous method applied to length scales near or above this critical length requires accounting for diffusion-coupling of spatial and temporal scales.

This technique also provides a framework for future research to evaluate how to best account for coupling of reaction and diffusion processes. It provides a good basis for examining the amount of this coupling, by identifying the diffusion-modulation of reaction eigenvalues. The method also has the ability to identify the slow dynamics of a reaction-diffusion system. When identified, the slow dynamics provide an ideal basis for reduction. This technique is easily extended to capture a larger number of spatial modes, allowing this method to be applied to globally analyze systems of longer characteristic lengths.

Acknowledgements

The authors recognize the support of the National Science Foundation (NSF) under Grant No. CBET-0650843.

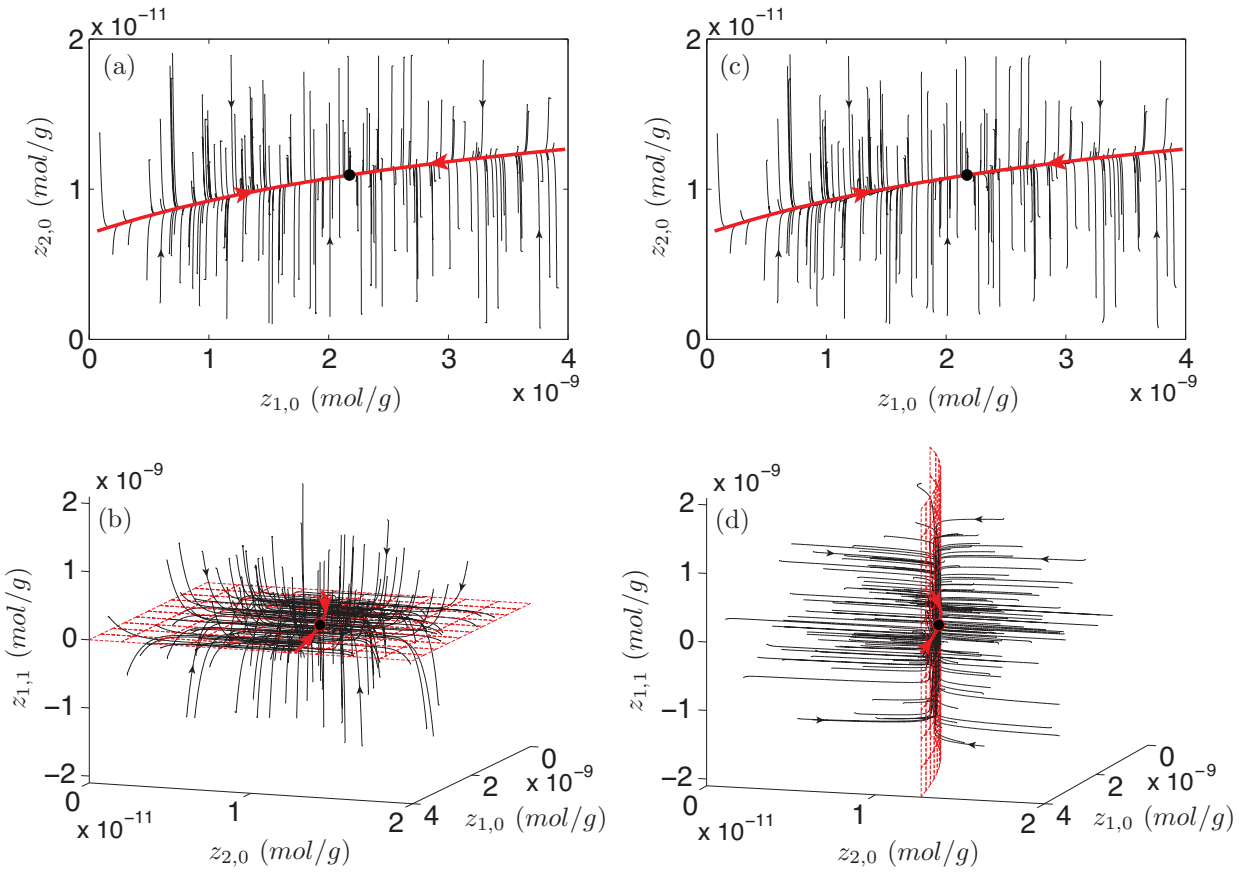

Figure 5: Projections of phase space for the Galerkin method of the Zel'dovich mechanism at $L = L_1$ (a,b) and $L = L_c$ (c,d), with random trajectories that have identical initial conditions when scaled.

References

[1] J.F. Griffith, Prog. Energy Combust. Sci. 21 (1) (1995) 25-107.

[2] T.F. Lu and C.K. Law, Prog. Energy Combust. Sci. 35 (2) (2009) 192-215.

[3] S. Singh, J.M. Powers, S. Paolucci, J. Chem. Phys. 117 (4) (2002) 1482-1496.

[4] Z. Ren and S.B. Pope, Combust. Theo. Model. 11 (5) (2007) 715-739.

[5] M.J. Davis, J. Phys. Chem. A. 110 (16) (2006) 5235-5256.

[6] M.J. Davis, J. Phys. Chem. A. 110 (16) (2006) 5257-5272.

[7] V. Bykov and U. Maas, Combust. Theor. Model. 11 (6) (2007) 839-862.

[8] S.H. Lam, Combust. Sci. Technol. 179 (4) (2007) 767-786.

[9] A. Adrover, F. Creta, M. Giona, and M. Valorani, J. Comput. Phys. 225 (2) (2007) 1442-1471.

[10] D.A. Goussis, M. Valorani, F. Creta, and H.N. Najm, Prog. Comput. Fluid Dyn. 5 (6) (2005) 316-326.

[11] M.J. Davis and R.T. Skodje, J. Chem. Phys. 111 (3) (1999) 859-874.

[12] A.N. Al-Khateeb, J.M. Powers, S. Paolucci, A.J. Sommese, J.A. Diller, J.D. Hauenstein, and J.D. Mengers, J. Chem. Phys. 131 (2) (2009) 024118.

[13] J. Robinson, Infinite-Dimensional Dynamical Systems, Cambridge University Press, Cambridge, U.K., 2001, p. 197-198, 371.

[14] R.J. Kee, L.G. Dixon, J. Warnatz, M.E. Coltrin, and J.A. Miller, A Fortran computer code package for the evaluation of gas-phase multicomponent transport properties, Report No. SAND86-8246, Sandia National Laboratories, 1991.