Chemical Engineering Science 65 (2010) 5429–5441

Contents lists available at ScienceDirect

Chemical Engineering Science

journal homepage: www.elsevier.com/locate/ces

Comparison of the stochastic fields method and DQMoM-IEM as turbulent reaction closures

Jethro Akroyd, Alastair J. Smith, Laurence R. McGlashan, Markus Kraft *

Department of Chemical Engineering and Biotechnology, University of Cambridge, New Museums Site, Pembroke Street, Cambridge CB2 3RA, UK

ARTICLE INFO

Article history:
Received 8 April 2010
Received in revised form
21 June 2010
Accepted 30 June 2010
Available online 7 July 2010

Keywords:
Simulation
Chemical reactors
Turbulence
CFD
DQMoM-IEM
SF

ABSTRACT

This paper compares two mean reaction rate closures for turbulent reacting flow: the Stochastic Fields (SF) method and the Direct Quadrature Method of Moments using the Interaction by Exchange with the Mean micromixing model (DQMoM-IEM). The methods have many common features and have received significant attention in recent literature, yet have not been systematically compared. We present both methods in the same mathematical framework and compare their numerical performance. In addition, we introduce antithetic sampling as a variance reduction technique to increase the efficiency of the SF algorithm. We extend the methodology to take advantage of this development and show details of the implementation of each method in a commercial computational fluid dynamics code. We present a systematic investigation and consider both axisymmetric and 3D formulations of a problem known from the literature. DQMoM-IEM showed excellent agreement with experimental and transported probability density function data. SF gave reasonable agreement, but retained a minor grid-dependence not seen with DQMoM-IEM and did not fully resolve the sub-grid segregation of the species. The antithetic sampling was demonstrated to significantly increase the efficiency of the axisymmetric SF cases.

1. Introduction

Turbulent reacting flows are a key field of research for many engineering applications. For example, the production of titanium dioxide from titanium tetrachloride and oxygen. Current turbulent flow methods separate the velocity and scalar quantities (such as species concentration) into resolved and unresolved components. For example, Reynolds-Averaged Navier Stokes (RANS) methods solve transport equations for average components, but need to close terms arising from unresolved fluctuating components. In reacting flows, the *chemical source terms* that occur in the material and energy balance equations are left unclosed and must be modelled.

Many approaches to modelling turbulent reacting flow have been studied (Libby and Williams, 1980; Peters, 2000; Fox, 2003). Transported probability density function (PDF) methods (Haworth, 2010) are applicable to all flows and offer the key advantage that the chemical source term does not need to be closed. However, Monte Carlo solution techniques are typically required (Pope, 1985). These may be computationally expensive and not necessarily suited to the computational fluid dynamics (CFD) software that would often be the method of choice for turbulent flow simulations.

Two methods considered in recent literature and amenable to implementation within existing CFD codes are the Direct Quadrature Method of Moments using the Interaction by Exchange with the Mean micromixing model (DQMoM-IEM) and the Stochastic Fields (SF) method. Despite the attention, no studies have directly compared the methods.

The application of DQMoM-IEM to turbulent reacting flows was suggested by Fox (2003). A joint composition PDF transport equation is approximated using a weighted discretisation in composition space and the interaction by exchange with the mean (IEM) micromixing model (Villermaux and Devillon, 1972). An arbitrary moment set is used to derive transport equations for the approximated PDF that guarantee to reproduce the correct transport of the moments within this set. The method has been applied to reactive precipitation in non-ideal plug flow reactors (Wang and Fox, 2004), nano-particle precipitation in confined impinging jet reactors (Raman et al., 2006; Lui and Fox, 2006, Gavi et al., 2007a, b) and stabilised turbulent methane-hydrogen flames (Tang et al., 2007).

The SF method (Valiño, 1998; Sabel'nikov and Soulard, 2005) approximates a joint composition PDF transport equation using a set of Eulerian 'stochastic fields'. The fields are defined over the

* Corresponding author. Tel.: +44 1223 762784; fax: +44 1223 334796.
E-mail address: mk306@cam.ac.uk (M. Kraft).
URL: http://como.cheng.cam.ac.uk (M. Kraft).

0009-2509/$ - see front matter © 2010 Elsevier Ltd. All rights reserved.
doi:10.1016/j.ces.2010.06.039

entire spatial domain and evolve according to a stochastic partial differential equation (SPDE) such that they remain statistically equivalent to a one-point joint composition PDF. The method has been applied to a number of turbulent reacting flows including the dispersion of reactive pollutants (Garmory et al., 2006, 2009), piloted methane flames (Mustata et al., 2006), and the auto-ignition of hydrogen and *n*-heptane flames (Jones et al., 2007; Jones and Navarro-Martinez, 2007, 2009).

The *purpose of this paper* is to compare the numerical behaviour of DQMoM-IEM and SF and to introduce antithetic sampling to improve the SF method. Section 2 introduces the key aspects of each method. Section 3 summarises the implementation of DQMoM-IEM and SF and explains the use of antithetic sampling to increase the efficiency of the SF simulations. Section 4 investigates the performance of DQMoM-IEM and SF against the method of moments and a turbulent reaction test case known from the literature (Li and Toor, 1986; Tsai and Fox, 1994; Tsai et al., 2002). Both axisymmetric and 3D cases are considered. The benefits of antithetic sampling are discussed and areas for further research suggested.

2. Theoretical background

This section introduces key aspects of the DQMoM-IEM and SF methods and discusses some common features. The derivations of the methods are documented in the literature and solve approximations to a closed joint composition PDF transport equation such as

$$\frac{\partial f_\phi}{\partial t}+\langle U_i\rangle\frac{\partial f_\phi}{\partial x_i}-\frac{\partial}{\partial x_i}\left(\Gamma_T\frac{\partial f_\phi}{\partial x_i}\right) = -\frac{\partial}{\partial \psi_\alpha}\left(\left[\frac{C_\phi}{2\tau_\phi}(\langle\phi_\alpha\rangle-\psi_\alpha)+S_\alpha(\psi)\right]f_\phi\right). \quad (1)$$

2.1. DQMoM-IEM

DQMoM-IEM was first derived by Fox (2003, Appendix B). This paper considers the case when the closed joint composition PDF transport Eq. (1) is approximated using

$$f_\phi(\psi;x,t)=f_\phi(\psi_1,\psi_2,\ldots,\psi_K;x,t) \approx \sum_{n=1}^{N} w^{(n)}(x,t)\prod_{\alpha=1}^{K}\delta_{\psi_\alpha^{(n)};x,t}, \quad (2)$$

where

$$\delta_{\psi_\alpha^{(n)};x,t}\equiv\delta[\psi_\alpha-\psi_\alpha^{(n)}(x,t)]. \quad (3)$$

The approximation introduces N weights $w^{(n)}$ and NK scalar composition variables $\psi_\alpha^{(n)}$, where $\alpha=1,\ldots,K$ scalars.

Transport equations that share the form of standard scalar transport equations are derived for the weights $w^{(n)}$ and weighted compositions $s_\alpha^{(n)}$:

$$\frac{\partial w^{(n)}}{\partial t}+\langle U_i\rangle\frac{\partial w^{(n)}}{\partial x_i}-\frac{\partial}{\partial x_i}\left(\Gamma_T\frac{\partial w^{(n)}}{\partial x_i}\right)=a^{(n)}, \quad (4)$$

$$\frac{\partial s_\alpha^{(n)}}{\partial t}+\langle U_i\rangle\frac{\partial s_\alpha^{(n)}}{\partial x_i}-\frac{\partial}{\partial x_i}\left(\Gamma_T\frac{\partial s_\alpha^{(n)}}{\partial x_i}\right)=b_\alpha^{(n)}, \quad (5)$$

where

$$s_\alpha^{(n)}\equiv w^{(n)}\psi_\alpha^{(n)}. \quad (6)$$

The source terms $a^{(n)}$ are set to zero and the weights evolve as conserved scalars subject to Eq. (4). A set of $M=NK$ unmixed empirical moments

$$\langle\phi_\alpha^{m_{\lambda\alpha}}\rangle_N=\sum_{n=1}^{N} w^{(n)}\psi_\alpha^{(n)m_{\lambda\alpha}} \quad \text{for } \lambda=1,\ldots,M \quad (7)$$

are used to derive a linear system of NK equations for the source terms $b_\alpha^{(n)}$. Note that these are *empirical* moments because they are moments of the approximated PDF as opposed to the true PDF. In the case that the unmixed empirical moments are specified

$$m_{\lambda\alpha}=\lambda \quad \text{for } \lambda=1,\ldots,N \text{ and } \alpha=1,\ldots,K, \quad (8)$$

the linear system can be solved to give a set of N equations for each scalar $\alpha=1,\ldots,K$ (Akroyd et al., 2010)

$$b_\alpha^{(n)}=b_{\mathrm{mx}_\alpha}^{(n)}+b_{\mathrm{rx}_\alpha}^{(n)}+b_{\mathrm{dx}_\alpha}^{(n)}, \quad (9)$$

where

$$b_{\mathrm{mx}_\alpha}^{(n)}=w^{(n)}\frac{C_\phi}{2\tau_\phi}(\langle\phi_\alpha\rangle_N-\psi_\alpha^{(n)}), \quad (10)$$

$$b_{\mathrm{rx}_\alpha}^{(n)}=w^{(n)}S_\alpha(\psi^{(n)}), \quad (11)$$

$$b_{\mathrm{dx}_\alpha}^{(n)}=w^{(n)}c_{\alpha\alpha}^{(n)}\sum_{\substack{i=1\\ i\neq n}}^{N}\frac{1}{\psi_\alpha^{(n)}-\psi_\alpha^{(i)}} +\prod_{\substack{i=1\\ i\neq n}}^{N}\frac{1}{\psi_\alpha^{(n)}-\psi_\alpha^{(i)}}\sum_{\substack{j=1\\ j\neq n}}^{N} w^{(j)}c_{\alpha\alpha}^{(j)}\prod_{\substack{k=1\\ k\neq j,n}}^{N}(\psi_\alpha^{(j)}-\psi_\alpha^{(k)}), \quad (12)$$

and

$$c_{\alpha\beta}^{(n)}\equiv\Gamma_T\frac{\partial\psi_\alpha^{(n)}}{\partial x_i}\frac{\partial\psi_\beta^{(n)}}{\partial x_i}. \quad (13)$$

The $b_{\mathrm{mx}_\alpha}^{(n)}$ and $b_{\mathrm{rx}_\alpha}^{(n)}$ terms describe micromixing and chemical reaction, $b_{\mathrm{dx}_\alpha}^{(n)}$ describes the effect of turbulent diffusion in the presence of spatial gradients of scalar α. We refer to them as the DQMoM-IEM *micromixing*, *reaction* and *diffusion* terms. The diffusion term is poorly conditioned and is singular if any $\psi_\alpha^{(n)}$ are equal. Its numerical treatment is investigated in detail by Akroyd et al. (2010).

2.2. SF

The SF method was derived independently by Valiño (1998) and Sabel'nikov and Soulard (2005). The derivations differ in that Valiño derives an Itô SPDE under the restriction that the fields are twice differentiable in space, whereas Sabel'nikov and Soulard present a more general derivation of a Stratonovich SPDE and show equivalence to Valiño's result.

This paper considers the case of a multivariate Itô SPDE, first given by Valiño and Hauke and Valiño (2004). The derivation follows the approach outlined by Valiño (1998). The joint composition PDF transport Eq. (1) is approximated using an ensemble of N fields

$$f_\phi(\psi;x,t)=f_\phi(\psi_1,\psi_2,\ldots,\psi_K;x,t) \approx \frac{1}{N}\sum_{n=1}^{N}\prod_{\alpha=1}^{K}\delta_{\psi_\alpha^{(n)};x,t}, \quad (14)$$

where $\delta_{\psi_\alpha^{(n)};x,t}$ is defined as per Eq. (3). Hauke and Valiño (2004) state the equivalent SPDE describing the transport of each field $\psi_\alpha^{(n)}(x,t)$:

$$\mathrm{d}\psi_\alpha^{(n)}=-\langle U_i\rangle\frac{\partial\psi_\alpha^{(n)}}{\partial x_i}\,\mathrm{d}t+\frac{\partial}{\partial x_i}\left(\Gamma_T\frac{\partial\psi_\alpha^{(n)}}{\partial x_i}\right)\mathrm{d}t +\frac{C_\phi}{2\tau_\phi}(\langle\phi_\alpha\rangle_N-\psi_\alpha^{(n)})\,\mathrm{d}t+S_\alpha(\psi^{(n)})\,\mathrm{d}t +(2\Gamma_T)^{1/2}\frac{\partial\psi_\alpha^{(n)}}{\partial x_i}\,\mathrm{d}W_i^{(n)}, \quad (15)$$