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A filtered mass density function approach for modeling separated two-phase flows for LES I: Mathematical formulation

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Abstract

The overall objective of this study is to develop a full velocity-scalar filtered mass density function (FMDF) formulation for large eddy simulation (LES) of a separated two-phase flow. Required in the development of the two-phase FMDF transport equation are the local instantaneous equations of motion for a two-phase flow previously derived by Kataoka. In Kataoka's development, phase interaction terms are cast in terms of a Dirac delta distribution on the phase interface. For this reason, it is difficult to close these coupling terms in the instantaneous formulation and this difficulty is propagated into the phase-coupling terms in the FMDF transport equation. To address this point a new derivation of the local instantaneous equations for a separated two-phase flow is given. The equations are shown to be consistent with the formulation given by Kataoka, and in the development, a direct link between the conditionally surface-filtered coupling terms, arising in the FMDF formulation, and LES phase-coupling terms is established. Clarification of conditions under which conditionally filtered interphase conversion terms in the marginal FMDF transport equations may be disregarded in a separated continuum-dispersed phase flow is discussed. Modeling approaches and solutions procedures to solve the two-phase FMDF transport equation via Monte-Carlo methods are outlined.
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1. Introduction

A multiphase flow may be defined as a flow field composed of a number of physically distinct and separate material phases together in one flow domain. Material phases can be non-miscible, or miscible depending on the existence and type of reaction mechanism present in a given material. Mathematical modeling of multiphase flows is challenging and several approaches have been taken in the past with varying degrees of success.

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One of the earliest models for multiphase flows is the continuum mixture model (Zuber and Findley, 1965; Truesdell, 1984). Two-phase mixture theory assumes that in a very small volume of the mixture, both phases of the flow co-exist. This approach is useful if one phase is finely dispersed in the other, however, due to this limiting assumption, predictions using mixture theory deviate from reality when the size of the dispersed phase approaches that of the carrier phase (Kataoka, 1986).

An alternative approach is the separated flow model. In this approach it is assumed that each phase can be treated separately with a moving boundary describing the phase interface. Local instantaneous conservation equations can be written down for each phase with jump conditions formulated across interphase boundaries to form a complete field description (Kataoka, 1986; Banerjee and Chan, 1980; Drew and Passman, 1998). In principle the local and instantaneous field equations can be derived exactly. The solution of these equations, however, is an extremely difficult and computationally time consuming task even for the simplest geometries and flow field configurations. Fortunately, for many engineering calculations an adequate amount of the salient physics of a system may be extracted from gross flow field features, such as mean temperatures, pressures, etc. therefore, mean-field-theory approaches are attractive alternatives to direct numerical simulations of the instantaneous equations. A mean-field theory in this sense represents any reduced description of the field that simplifies the equations of motion at the cost of "smearing-out" the detailed information contained in the original equations. Traditional approaches for separated two-phase flow rely on an averaging procedure of some kind viz., time, space or ensemble (Banerjee and Chan, 1980; Grey and Lee, 1976; Drew and Passman, 1998). In this study, the definition of the phase-filtered equations for large eddy simulation (LES), and consequently the notion of the average itself, are based on spatial filtering; hence, the equations in the context are referred to as phase-filtered. It should be noted that alternative interpretations of the phase-averaged equations are available based on either time or ensemble averaging (Drew and Passman, 1998). The choice of spatial averaging here is chosen to be consistent with the enormous body of LES literature that is based on spatial averaging techniques. The phase-average defined in this context is an approximation of the ensemble average, and is a valid estimation of the mathematical expectation within the approximations already taken in the LES formulation. One disadvantage of a phase-averaged approach, however, is that moment closures, necessitated from reducing the number of degrees of freedom in the field, often inadequately capture the effects of physics at the small scales.

A promising approach for modeling single-phase turbulent chemically reactive flows are probability density function (PDF) and filtered density function (FDF) methods that have been developed over the last 30 years (Lundgren, 1967; Pope, 1985, 1976; Dopazo and O'Brien, 1976; O'Brien, 1980; Rhodes, 1975; Bilger et al., 1976; Janicka et al., 1979; Givi, 1989). The relative advantages and disadvantages of PDF and FDF methods for single-phase turbulent chemically reactive flows have been well documented in the literature (Pope, 1985; Colucci et al., 1998; Zhou and Pereira, 2000; Jaberi, 1999; Givi, 2003; Fox, 2003). A current area of research is the application of PDF and FDF methods for two-phase flows formulated either for full velocity-scalar PDF (full PDF) of both phases (Minier and Peirano, 2001), or scalar-PDF/FDF of both phases coupled to traditional Reynolds averaged Navier–Stokes (RANS) or LES of the momentum fields. Recently, Zhu et al. (2000) have formulated and solved a two-phase flow consisting of reactive droplets dispersed in a turbulent carrier gas with a PDF–RANS formulated gas field (PDF formulated scalar field coupled to RANS formulated momentum field) and Williams spray equation (Williams, 1958) describing the droplet phase. Zhu's work confirms the feasibility of a PDF approach for spray combustion applications. Minier and Peirano (2001) have applied the PDF method for the simulation of turbulent polydispersed two-phase flows.

To the authors' knowledge, there has been little attention paid to date in the area of LES–FDF formulations for two-phase flows. The objective of this current research is to present a FDF formulation for a two-phase flow that is consistent with standard LES formulations of two-phase flows based on spatial filtering. It will be shown that in order to derive the two-phase FDF transport equation, the local instantaneous equations for two-phase flow are required. Attempts at deriving the FDF transport equation by simply "plugging-in" the instantaneous equations of Kataoka (1986) result in conditionally surface-filtered phase-coupling terms that are awkward to interpret and consequently impossible to model with any rigor. Namely, terms of the form: $\langle Qa^I|_{surface}\rangle$, representing the conditional surface filter of a field quantity Q multiplied by the "interfacial area concentration", a^I, naturally fall out of both the PDF formulation as derived and discussed by Zhu et al. (2000) and the FDF formulation given herein. The challenge in modeling these conditionally surface-filtered

terms is the interpretation of a^I; viz., Kataoka (1986) defined a^I as a Dirac delta distribution of an interface function separating the two phases of the flow. To the authors' knowledge, previous attempts at deriving the PDF transport equation in the context of ensemble averaging do not attempt to resolve this issue; rather, a phenomenological PDF spray equation is simply given (Zhu et al., 2000, 1996; Rumberg and Rogg, 2000). In this study, the interpretation of the conditionally surface-filtered coupling terms is considered from first principles in the context of LES. A key step in the analysis is a new definition and interpretation of a^I based on a new method of deriving the instantaneous separated two-phase flow equations. With this new definition of a^I, conditionally surface-filtered terms can be cast in terms of filtered surface terms that are consistent with standard LES formulations of two-phase flows. Models for these filtered surface terms are often known for a given application, therefore, one may use the phase-filtered coupling terms in the LES equations as a guide for postulating closures for the conditionally surface-filtered coupling terms arising in the two-phase FDF formulation.

The remainder of this article is organized as follows: in Section 2 the field equations for a reactive flow are summarized. In Section 3 the equations in Section 2 are spatially filtered to arrive at the phase-filtered LES equations for a separated two-phase flow. In Section 4 the LES equations of Section 3 are considered in the limit where the filtering kernel approaches a Dirac delta distribution. The motivation for taking this limit, as mentioned above, is to arrive at a new definition of the instantaneous interfacial area concentration, a^I, that will be used in Section 6 to simplify phase-coupling terms. In Section 5 the two-phase FDF transport equation is derived. In Section 6, phase-coupling and interphase volumetric conversion terms in the context of the probabilistic formulation are analyzed and discussed. Section 7 summarizes the major findings of this study.

2. Single phase conservation equations

The equations governing the reactive flow for a Newtonian flow field may be written as

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{v}) = 0 \tag{1}$$

$$\frac{\partial}{\partial t}(\rho \mathbf{v}) + \nabla \cdot (\rho \mathbf{v} \otimes \mathbf{v}) = \nabla \cdot \underset{\sim}{\boldsymbol{\sigma}} + \rho \mathbf{f} \tag{2}$$

$$\frac{\partial}{\partial t}(\rho Y_\beta) + \nabla \cdot (\rho \mathbf{v} Y_\beta) = -\nabla \cdot (\mathbf{J}_\beta) + \rho S_\beta \tag{3}$$

$$\frac{\partial}{\partial t}(\rho h) + \nabla \cdot (\rho \mathbf{v} h) = -\nabla \cdot (\mathbf{J}_h) + \rho S_h \tag{4}$$

representing the conservation of mass, momentum, species' mass fraction and sensible enthalpy, respectively. Implicit in Eq. (4) are the assumptions of ordinary diffusion and unity Lewis number. The effects of pressure work, viscous dissipation and reaction rates are contained in the term, S_h. Eqs. (1)–(4) along with appropriate constitutive models, compatibility constraints, equation of state and initial/boundary conditions, completely describe the continuum field given by ρ, $\mathbf{v}$, Y_β, and h. Furthermore, Eqs. (1)–(4) may be compactly expressed as

$$\frac{\partial}{\partial t}(\rho \psi) + \nabla \cdot (\rho \psi \mathbf{v}) = -\nabla \cdot \mathbf{J} + \rho S \tag{5}$$

by identification of the appropriate conserved quantity $\rho \psi$ with flux $\mathbf{J} \equiv \rho \mathbf{j} (= -\kappa \nabla T, -\rho \mathscr{D}_m \nabla Y_\beta$ or $-\mu/Sc \nabla h)$ and source S. In a separated two-phase flow, Eq. (5) is satisfied within each phase of the flow for some conserved quantity $\rho \psi$, and may be phase-averaged to relate averaged properties within one phase to those in the other phase.

3. Phase-filtered conservation equations

To derive the phase-filtered LES equations, Eq. (5) is filtered using a convolution integration with filtering kernel G. In the integration, G restricts the filtering operation to a compact region in the spatial domain of the flow. Let $G(\mathbf{x}' - \mathbf{x})$, defined for $\mathbf{x} > 0$, $t > 0$ be such that it is parity invariant, non-zero, continuously differentiable and normalized with compact support.

$$G(\mathbf{x}' - \mathbf{x}) = G(-\mathbf{x}' + \mathbf{x}) \tag{6}$$

$$G(\mathbf{x}' - \mathbf{x}) \geqslant 0 \quad \forall \mathbf{x} \in \mathbb{R}^3 \tag{7}$$

$$G(\mathbf{x}' - \mathbf{x}) \in C_0^\infty \tag{8}$$

$$\int_\infty \mathrm{d}^3 x' G(\mathbf{x}' - \mathbf{x}) = 1 \tag{9}$$

Using this kernel, the local phase-filtered volume fraction in phase 1 of a two-phase flow may be defined as

$$\theta_1(\mathbf{x}, t) \equiv \int_{\Omega_1} \mathrm{d}^3 x' G(\mathbf{x}' - \mathbf{x}) \tag{10}$$

where Ω_1 is the region occupied by material 1, Ω_2 is the region occupied by material 2 and the entire flow domain is given by $\Omega_T \equiv \Omega_1 + \Omega_2$. In the two-phase flow, θ_2 is specified in terms of $\theta_1 : \theta_2 = 1 - \theta_1$. For a separated two-phase flow both Ω_1 and Ω_2 are functions of t and are such that Ω_T is constant at any given spatial location and time. The phase-filtered value of a field property ψ_1 in phase 1 is defined as (Kuo, 1986; Grey and Lee, 1976)

$$\overline{\psi_1(\mathbf{x}, t)} \equiv \frac{1}{\theta_1(\mathbf{x}, t)} \int_{\Omega_1} \mathrm{d}^3 x' \psi_1(\mathbf{x}', t) G(\mathbf{x}' - \mathbf{x}) \tag{11}$$

where $\overline{\psi}$ and $\langle \psi \rangle \equiv \theta \overline{\psi}$ are the intrinsic and extrinsic phase filtered values, respectively. When filtering Eq. (5) according to Eq. (11), a key point is that the filtering operation does not commute with the time derivative operator so that Leibnitz's rule must be used (Grey and Lee, 1976),

$$\frac{\partial}{\partial t} \int_{\Omega_k(t)} \mathrm{d}^3 x' G \rho_k \psi_k = \int_{\Omega_k(t)} \mathrm{d}^3 x' \frac{\partial}{\partial t} G \rho_k \psi_k + \oint_{\partial \Omega_k(t)} \mathrm{d}^2 x' G \rho_k \psi_k (\mathbf{v}^\mathrm{I} \cdot \mathbf{e}_{n,k}^\mathrm{I}) \tag{12}$$

where $\mathbf{v}^\mathrm{I}$ and $\mathbf{e}_{n,k}^\mathrm{I}$ are the interface velocity and interface unit normal, oriented positive out of $\partial \Omega_k(t)$, respectively. Filtering Eq. (5) over phase k and utilizing Eqs. (10)–(12) results in the following phase-filtered LES equation for a separated two-phase flow:

$$\frac{\partial}{\partial t} \left(\theta_k \overline{\rho_k \psi_k} \right) + \nabla \cdot \left(\theta_k \overline{\rho_k \mathbf{j}_k} \right) + \nabla \cdot \left(\theta_k \overline{\rho_k \psi_k \mathbf{v}_k} \right) - \theta_k \overline{\rho_k S_k}$$
$$= -\oint_{\partial \Omega_k} \mathrm{d}^2 x' G \rho_k \psi_k (\mathbf{v}_k - \mathbf{v}^\mathrm{I}) \cdot \mathbf{e}_{n,k}^\mathrm{I} - \oint_{\partial \Omega_k} \mathrm{d}^2 x' G \rho_k \mathbf{j}_k \cdot \mathbf{e}_{n,k}^\mathrm{I} \tag{13}$$

For a variable density flow, it is advantageous to cast Eq. (13) in terms of Favre filtered quantities defined as

$$\tilde{\beta} \equiv \frac{\overline{\beta \rho}}{\overline{\rho}} \Rightarrow \overline{\beta \rho} = \overline{\rho} \tilde{\beta} \tag{14}$$

and subsequently a subgrid scale (SGS) correlation $\mathscr{T}_k$ may be defined as

$$\mathscr{T}_k \equiv \theta_k \overline{\rho}_k \tilde{\psi}_k \tilde{\mathbf{v}}_k - \theta_k \overline{\rho_k \psi_k \mathbf{v}_k} \tag{15}$$

where the (nonlinear) correlation quantity $\mathscr{T}_k$ accounts for the effect of unresolved turbulent transport of ψ in phase k. Expressing Eq. (13) in terms of Favre filtered quantities yields,

$$\frac{\partial}{\partial t} (\theta_k \overline{\rho}_k \tilde{\psi}_k) + \nabla \cdot (\theta_k \overline{\rho}_k \tilde{\psi}_k \tilde{\mathbf{v}}_k) - \nabla \cdot \mathscr{T}_k + \nabla \cdot (\theta_k \overline{\rho}_k \tilde{\mathbf{j}}_k) - \theta_k \overline{\rho}_k \tilde{S}_k$$
$$= -\oint_{\partial \Omega_k} \mathrm{d}^2 x' G \rho_k \psi_k (\mathbf{v}_k - \mathbf{v}^\mathrm{I}) \cdot \mathbf{e}_{n,k}^\mathrm{I} - \oint_{\partial \Omega_k} \mathrm{d}^2 x' G \rho_k \mathbf{j}_k \cdot \mathbf{e}_{n,k}^\mathrm{I} = -\oint_{\partial \Omega_k} \mathrm{d}^2 x' G (\psi_k \dot{m}_k'' + \rho_k \mathbf{j}_k \cdot \mathbf{e}_{n,k}^\mathrm{I}) \tag{16}$$

The r.h.s. of Eq. (16) represents phase-coupling terms (PCT) for the quantity ψ_k where $\dot{m}_k'' \equiv \rho_k (\mathbf{v}_k - \mathbf{v}^\mathrm{I}) \cdot \mathbf{e}_{n,k}^\mathrm{I}$ is the interphase mass transfer rate. The PCT's may be more compactly written by introducing the definition of a local surface-filtered field property filtered in phase k as (Kuo, 1986),

$$\overline{(\psi_k(\mathbf{x}, t))_s} \equiv \frac{\oint_{\partial \Omega_k} \mathrm{d}^2 x' \psi_k(\mathbf{x}', t) G(\mathbf{x}' - \mathbf{x})}{\oint_{\partial \Omega_k} \mathrm{d}^2 x' G(\mathbf{x}' - \mathbf{x})} \tag{17}$$

Furthermore, the denominator of Eq. (17) representing the filtered interface surface area per unit volume is given by Kuo (1986)

$$\oint_{\partial \Omega_k} \mathrm{d}^2 x' G(\mathbf{x}' - \mathbf{x}) = \frac{\theta_k A_k}{V_k} \tag{18}$$

where A_k and V_k are the area and volume occupied by phase k at time t. Thus, the PCT on the r.h.s. of Eq. (16) may then be written as

$$\begin{aligned} \mathrm{PCT} &= -\oint_{\partial \Omega_k} \mathrm{d}^2 x' G(\psi_k \dot{m}_k'' + \rho_k \mathbf{j}_k \cdot \mathbf{e}_{n,k}^{\mathrm{I}}) = -\left(\overline{\psi_k \dot{m}_k''} + \overline{\rho_k \mathbf{j}_k \cdot \mathbf{e}_{n,k}^{\mathrm{I}}}\right)_s \oint_{\partial \Omega_k} \mathrm{d}^2 x' G(\mathbf{x} - \mathbf{x}') \\ &= -\left(\overline{\psi_k \dot{m}_k''} + \overline{\rho_k \mathbf{j}_k \cdot \mathbf{e}_{n,k}^{\mathrm{I}}}\right)_s \frac{\theta_k A_k}{V_k} \end{aligned} \tag{19}$$

Using Eq. (19), the phase-filtered LES transport equation for the conserved quantity ψ over phase k is written as

$$\frac{\partial}{\partial t}(\theta_k \overline{\rho}_k \widetilde{\psi}_k) + \nabla \cdot (\theta_k \overline{\rho}_k \widetilde{\psi}_k \widetilde{\mathbf{v}}_k) - \nabla \cdot \mathscr{T}_k + \nabla \cdot (\theta_k \overline{\rho}_k \overline{\mathbf{j}}_k) - \theta_k \overline{\rho}_k \widetilde{S}_k = -\left(\overline{\psi_k \dot{m}_k''} + \overline{\rho_k \mathbf{j}_k \cdot \mathbf{e}_{n,k}^{\mathrm{I}}}\right)_s \frac{\theta_k A_k}{V_k} \tag{20}$$

Appropriately substituting for ψ_k into Eq. (20) for mass, momentum, species and sensible enthalpy conservation, results in the following phase-filtered two-phase flow equations:

$$\frac{\partial}{\partial t}(\theta_k \overline{\rho}_k) + \nabla \cdot (\theta_k \overline{\rho}_k \widetilde{\mathbf{v}}_k) = -(\overline{\dot{m}_k''})_s \frac{\theta_k A_k}{V_k} \tag{21}$$

$$\frac{\partial}{\partial t}(\theta_k \overline{\rho}_k \widetilde{\mathbf{v}}_k) + \nabla \cdot (\theta_k \overline{\rho}_k \widetilde{\mathbf{v}}_k \otimes \widetilde{\mathbf{v}}_k) - \nabla \cdot \underset{\sim}{\mathscr{T}}_k^{(v)} - \nabla \cdot (\theta_k \underset{\sim}{\overline{\boldsymbol{\sigma}}}_k) - \theta_k \overline{\rho}_k \widetilde{\mathbf{f}}_k = -\left(\overline{\mathbf{v}_k \dot{m}_k''} + \underset{\sim}{\overline{\boldsymbol{\sigma}}_k \cdot \mathbf{e}_{n,k}^{\mathrm{I}}}\right)_s \frac{\theta_k A_k}{V_k} \tag{22}$$

$$\begin{aligned} &\frac{\partial}{\partial t}(\theta_k \overline{\rho}_k \widetilde{Y}_{\beta,k}) + \nabla \cdot (\theta_k \overline{\rho}_k \widetilde{Y}_{\beta,k} \widetilde{\mathbf{v}}_k) - \nabla \cdot \mathscr{T}_k^{(Y)} - \nabla \cdot (\theta_k \overline{\rho}_k \overline{\mathscr{D}}_{\beta,k} \nabla \widetilde{Y}_{\beta,k}) - \theta_k \overline{\rho}_k \widetilde{S}_{\beta,k} \\ &= -\left(\overline{Y_{\beta,k} \dot{m}_k'} + \overline{\rho_k \mathscr{D}_{\beta,k} \nabla Y_{\beta,k} \cdot \mathbf{e}_{n,k}^{\mathrm{I}}}\right)_s \frac{\theta_k A_s}{V_k} \end{aligned} \tag{23}$$

$$\begin{aligned} &\frac{\partial}{\partial t}(\theta_k \overline{\rho}_k \widetilde{h}_k) + \nabla \cdot (\theta_k \overline{\rho}_k \widetilde{h}_k \widetilde{\mathbf{v}}_k) - \nabla \cdot \mathscr{T}_k^{(h)} - \nabla \cdot \left(\frac{\theta_k \mu_k}{Sc_k} \nabla \widetilde{h}_k\right) - \theta_k \overline{\rho}_k \widetilde{S}_{h,k} \\ &= -\left(\overline{h_k \dot{m}_k''} + \overline{\frac{\mu_k}{Sc_k} \nabla h_k \cdot \mathbf{e}_{n,k}^{\mathrm{I}}}\right)_s \frac{\theta_k A_k}{V_k} \end{aligned} \tag{24}$$

The terms on the r.h.s. of the equalities in Eqs. (21)–(24) are the phase-filtered phase-coupling terms. These terms contain the physics of the interaction between phases and must be modeled to close the field. Closure of these terms is problem specific, but often known (Kuo, 1986; Whitaker, 1973; Gough and Zwarts, 1979). The above system of equations is closed when the PCT's, Favre filtered source terms and two-phase SGS correlations are known. Traditional methods for solving Eqs. (21)–(24) require closure models for the SGS physics contained in $\nabla \cdot \mathscr{T}_k^{\alpha}, \nabla \cdot \mathscr{T}_k^{(v)}, \widetilde{S}_{\beta,k}$ and $\widetilde{S}_{h,k}$. In moment closure approaches, it is assumed that most of the physical information contained in these nonlinear source terms can be expressed in terms of lower order moments, viz., means and variances (Wilcox, 1994). It is well known that this approach is acceptable for simple single-phase non-reacting flows but quickly becomes problematic for reacting and/or multiphase systems. For example, in a chemically reacting flow the reaction rate source terms in S_k may take an Arrhenius form which is highly nonlinear in the temperature T_k. In this case, it is not possible to model the mean of $\widetilde{S}_k$ using only means and variances of ρ_k and T_k separately without losing physical information contained in the reaction rate source term. An attractive alternative to traditional moment closures for turbulent chemically reacting flows are the probability density function (PDF) and filtered density function (FDF) methods pioneered by

Lundgren (1967), Dopazo (1994), Givi (1989) and Pope (1985), as well as others noted in Section 1. In PDF/FDF approaches, an equation governing the transport of PDF/FDF containing all the one-point statistical information of the flow field variables is developed and its solution is sought. One of the main advantages of PDF/FDF approaches is that reactive source terms are in closed form. In the next section the local instantaneous equations for a separated two-phase flow, needed in the FDF development of Section 5, are developed.

4. Local instantaneous two-phase conservation equations

The transport equation for the FDF is derived from the local instantaneous governing equations for the field. The starting point for development of the single-phase FDF equation is the set of instantaneous equations given in Eqs. (1)–(4) and compactly written in Eq. (5) (Givi, 1989; Pope, 2000; Gicquel et al., 2002). Analogously, the transport equation for the FDF of a two-phase flow must be derived from the local instantaneous two-phase flow equations. Motivated by measure-theoretic concepts, Kataoka (1986) developed the local instantaneous conservation equations for each phase of a separated two-phase flow. Kataoka used the theory of distributions to differentiate field quantities across the discontinuous phase interface in the separated field arriving at interface source terms defined in terms of a "local instantaneous interfacial area concentration", a^I, given in terms of a Dirac delta distribution. In the current study, the local instantaneous equations for each phase of a separated two-phase flow are derived starting directly from the phase-filtered equations given in Eqs. (21)–(24). It is found that the exact mathematical form of Kataoka's equations are recovered, including the local instantaneous interfacial area concentration term, by allowing the filtering kernel G to shrink to a Dirac delta distribution in the phase-filtered equations. Furthermore, a new interpretation of a^I is developed that will be useful in closing the two-phase FDF transport equation in Section 5.

In describing the instantaneous flow of two separated fluid phases, it is necessary to be able to distinguish between phases at a given location and instant in time. To this end, a characteristic function, $\gamma(\mathbf{x}, t)$, is introduced (Zhu, 1996) that describes the phase interface at a position $\mathbf{x}$ and time t

$$\gamma(\mathbf{x}, t) \begin{cases} < 0, & \text{in phase 1} \\ = 0, & \text{on the interface} \\ > 0, & \text{in phase 2} \end{cases} \tag{25}$$

A phase indicator function, denoted by $\phi(\mathbf{x}, t)$, may be constructed from $\gamma(\mathbf{x}, t)$ that physically represents the local instantaneous volume fraction at a position $\mathbf{x}$ and time t,

$$\phi_1(\mathbf{x}, t) \equiv 1 - H[\gamma(\mathbf{x}, t)] = \begin{cases} 1, & \gamma < 0 \text{ in phase 1} \\ 0, & \gamma > 0 \text{ in phase 2} \end{cases} \tag{26}$$

where H is the Heaviside function. Note, by construction $\phi_2 = 1 - \phi_1$. Requiring the streamline derivative of the phase indicator function to balance the interphase transition yields an equation of motion for $\phi_k(\mathbf{x}, t)$,

$$\frac{\partial \phi_k}{\partial t} + \mathbf{v}_k \cdot \nabla \phi_k = \Pi_k \tag{27}$$

where, Π_k describes the volume rate of change of phase k due to interphase conversion (Zhu, 1996). The exact mathematical form of this term is developed later in Section 6.2 (see Eq. (87)).

Consider the case for which G, defined by Eqs. (6)–(9), is specified as top-hat filter in terms of a Heaviside function $H(\mathbf{x})$. In one spatial dimension this can be written as

$$G\left(\frac{x' - x}{\Delta_f}\right) \equiv \frac{1}{\Delta_f}[H(x' - (x - \Delta_f/2)) - H(x' - (x + \Delta_f/2))] = \frac{1}{\Delta_f}[H(\eta + \Delta_f/2) - H(\eta - \Delta_f/2)] \tag{28}$$

where $\eta \equiv x' - x$, and Δ_f is the filter width in the context of LES. In the limit $\Delta_f \to 0, G$ approaches a Dirac delta distribution (Arfkin, 1985),

$$\lim_{\Delta_f \to 0} \frac{1}{\Delta_f}[H(\eta + \Delta_f/2) - H(\eta - \Delta_f/2)] = \delta(x' - x). \tag{29}$$

Using the property $\delta(\mathbf{x}) = \delta(x_1)\delta(x_2)\delta(x_3)\ldots$, one may write,

$$\lim_{\Delta_f \to 0} G\left(\frac{\mathbf{x}' - \mathbf{x}}{\Delta_f}\right) = \delta(\mathbf{x}' - \mathbf{x}). \tag{30}$$

Given the smoothness requirement in Eq. (8), the mean volume fraction in phase 1, given by Eq. (10), in this limit yields,

$$\lim_{\Delta_f \to 0} \theta_1(\mathbf{x}, t) = \lim_{\Delta_f \to 0} \int_{\Omega_1} \mathrm{d}^3 x' G\left(\frac{\mathbf{x}' - \mathbf{x}}{\Delta_f}\right) = \int_{\Omega_1} \mathrm{d}^3 x' \delta(\mathbf{x}' - \mathbf{x}) = \int_{\Omega_T} \mathrm{d}^3 x' \delta(\mathbf{x}' - \mathbf{x})\phi_1(\mathbf{x}', t) = \phi_1(\mathbf{x}, t) \tag{31}$$

which exactly recovers the instantaneous volume fraction defined earlier in Eq. (26). Thus, the result is that as the filter space reduces to a sample point, the local mean volume fraction recovers the local instantaneous volume fraction, i.e.,

$$\lim_{G \to \delta} \theta(\mathbf{x}, t) = \phi(\mathbf{x}, t) \tag{32}$$

Performing a similar operation on Eqs. (11) and (17) for $\overline{\psi_1}$ and $(\overline{\psi_1})_s$ results in,

$$\begin{aligned}
\lim_{\Delta_f \to 0} \overline{\psi_1(\mathbf{x}, t)} &= \lim_{\Delta_f \to 0} \left[\frac{1}{\theta_1(\mathbf{x}, t)} \int_{\Omega_1} \mathrm{d}^3 x' G\left(\frac{\mathbf{x}' - \mathbf{x}}{\Delta_f}\right)\psi_1(\mathbf{x}', t)\right] \\
&= \lim_{\Delta_f \to 0} \left[\frac{1}{\theta_1(\mathbf{x}, t)} \int_{\Omega_T} \mathrm{d}^3 x' G\left(\frac{\mathbf{x}' - \mathbf{x}}{\Delta_f}\right)\psi_1(\mathbf{x}', t)(1 - H[\gamma(\mathbf{x}', t)])\right] \\
&= \frac{1}{\phi_1(\mathbf{x}, t)} \int_{\Omega_T} \mathrm{d}^3 x' \delta(\mathbf{x}' - \mathbf{x})\psi_1(\mathbf{x}', t)(1 - H[\gamma(\mathbf{x}', t)]) \\
&= \frac{1}{\phi_1(\mathbf{x}, t)} \psi_1(\mathbf{x}, t)\phi_1(\mathbf{x}, t) = \psi_1(\mathbf{x}, t).
\end{aligned} \tag{33}$$

and,

$$\lim_{\Delta_f \to 0} \overline{(\psi(\mathbf{x}, t))_s} = \lim_{\Delta_f \to 0} \frac{\oint_{\partial\Omega_1} \mathrm{d}^2 x' \psi_1(\mathbf{x}', t) G\left(\frac{\mathbf{x}' - \mathbf{x}}{\Delta_f}\right)}{\oint_{\partial\Omega_1} \mathrm{d}^2 x' G\left(\frac{\mathbf{x}' - \mathbf{x}}{\Delta_f}\right)} = \frac{\oint_{\partial\Omega_1} \mathrm{d}^2 x' \psi_1(\mathbf{x}', t)\delta(\mathbf{x}' - \mathbf{x})}{\oint_{\partial\Omega_1} \mathrm{d}^2 x' \delta(\mathbf{x}' - \mathbf{x})} \equiv \psi_1^{\mathrm{I}}(\mathbf{x}, t). \tag{34}$$

Thus, in the limit where the filtering volume goes to zero, phase-filtered functions are sampled at a point, recovering their local instantaneous value, and surface-filtered values are evaluated at the phase interface, where formally they are defined as

$$\psi_1^{\mathrm{I}} \equiv \lim_{\gamma \to -0} \psi_1 \tag{35}$$

$$\psi_2^{\mathrm{I}} \equiv \lim_{\gamma \to +0} \psi_2 \tag{36}$$

Considering the correlation $\mathscr{T}_k$, the result $\lim_{\Delta_f \to 0}(\overline{\rho}_k\tilde{\psi}_k) = \rho_k\psi_k$, yields,

$$\lim_{\Delta_f \to 0} \nabla \cdot \mathscr{T}_k = \nabla \cdot \lim_{\Delta_f \to 0} \mathscr{T}_k = \nabla \cdot \left[\lim_{\Delta_f \to 0}(\theta_k \overline{\rho}_k \tilde{\psi}_k \tilde{\mathbf{v}}_k) - \lim_{\Delta_f \to 0}(\theta_k \overline{\rho_k \psi_k \mathbf{v}_k})\right] = \phi_k \rho_k \psi_k \mathbf{v}_k - \phi_k \rho_k \psi_k \mathbf{v}_k = 0 \tag{37}$$

Thus, the correlation $\mathscr{T}_k$ vanishes as the filtering volume goes to zero. This is expected physically as the information associated with a spatial region around any fluid element goes to zero with Δ_f, and hence the correlation distance reduces to zero.

Lastly, a useful identity for a^{I} may be obtained by considering the denominator of Eq. (34) directly. Considering the general surface normal, $\mathbf{e}_{n,1}^{\mathrm{I}} = f(\mathbf{x})$, with integration over $\mathbf{x}'$ one may write

$$\oint_{\partial\Omega_1} \mathrm{d}^2 x' \delta(\mathbf{x}' - \mathbf{x}) = \mathbf{e}_{n,1}^{\mathrm{I}}(\mathbf{x}) \cdot \oint_{\partial\Omega_1} \mathrm{d}^2 x' \mathbf{e}_{n,1}^{\mathrm{I}}(\mathbf{x})\delta(\mathbf{x}' - \mathbf{x}). \tag{38}$$

Viewing the surface normal in the integrand of Eq. (38) as a test function, the gradient theorem (Arfkin, 1985) may be applied to Eq. (38) resulting in

$$\mathbf{e}_{n,1}^{\mathrm{I}} \cdot \oint_{\partial\Omega_1} \mathrm{d}^2\mathbf{x}'\delta(\mathbf{x}'-\mathbf{x}) = \mathbf{e}_{n,1}^{\mathrm{I}} \cdot \int_{\Omega_1} \mathrm{d}^3x'\nabla\delta(\mathbf{x}'-\mathbf{x}) = \mathbf{e}_{n,1}^{\mathrm{I}} \cdot \int_{\Omega_{\mathrm{T}}} \mathrm{d}^3x'\nabla\delta(\mathbf{x}'-\mathbf{x})\phi_1(\mathbf{x}',t)$$

$$= \mathbf{e}_{n,1}^{\mathrm{I}} \cdot \int_{\Omega_{\mathrm{T}}} \mathrm{d}^3x'\nabla\delta(\mathbf{x}'-\mathbf{x})(1-H[\gamma(\mathbf{x}',t)]) = -\mathbf{e}_{n,1}^{\mathrm{I}} \cdot \nabla(1-H[\gamma(\mathbf{x},t)])$$

$$= \mathbf{e}_{n,1}^{\mathrm{I}} \cdot \nabla H[\gamma(\mathbf{x},t)] = \mathbf{e}_{n,1}^{\mathrm{I}} \cdot \left\{\frac{\mathrm{d}H}{\mathrm{d}\gamma}\frac{\partial\gamma}{\partial\mathbf{x}}\right\} = \mathbf{e}_{n,1}^{\mathrm{I}} \cdot \nabla\gamma(\mathbf{x},t)\delta(\gamma) \tag{39}$$

where the definition of the Dirac delta, $\delta(x) \equiv \mathrm{d}H(x)/\mathrm{d}x$, has been used (Arfkin, 1985). Simple differential geometry dictates that the unit normal vector off the phase interface (oriented outward from phase 1 into phase 2) is given by

$$\mathbf{e}_{n,1}^{\mathrm{I}} = \frac{\nabla\gamma}{|\nabla\gamma|} \tag{40}$$

Substituting Eq. (40) into Eq. (39) yields

$$\oint_{\partial\Omega_1} \mathrm{d}^2x'\delta(\mathbf{x}'-\mathbf{x}) = \frac{\nabla\gamma}{|\nabla\gamma|} \cdot \nabla\gamma\delta(\gamma) = \frac{\nabla\gamma\cdot\nabla\gamma}{|\nabla\gamma|} = \frac{|\nabla\gamma|^2}{|\nabla\gamma|}\delta(\gamma) = |\nabla\gamma|\delta(\gamma). \tag{41}$$

The quantity $|\nabla\gamma|\delta(\gamma) \equiv a^{\mathrm{I}}$ is the interfacial area concentration consistent with that given by Kataoka (1986). If γ represents a level-set (Sethian, 1999), then $|\nabla\gamma| = 1$ and the interfacial area concentration is identically a Dirac delta distribution. Formally, then, it has been shown that

$$\lim_{\Delta_f\to0} \oint_{\partial\Omega_1} \mathrm{d}^2x'G(\mathbf{x}'-\mathbf{x}) = \oint_{\partial\Omega_1} \mathrm{d}^2x'\delta(\mathbf{x}'-\mathbf{x}) \equiv a^{\mathrm{I}} = \text{Interfacial Area Concentration} \tag{42}$$

The fact that a^{I} is shown to have the form given in Eq. (42) is of *key* importance in the two-phase FDF formulation. As will be discussed in Section 5, phase coupling arises in the FDF context in terms of conditionally filtered flux terms conditioned *on the phase interface*, including the instantaneous area concentration in the surface filtering operation. It will be shown in Section 6, using the result in Eq. (42), that these conditionally surface-filtered coupling terms, whose closure is necessary for any practical application of this theory, can be cast *directly* into the form of PCT's arising in the LES formulation shown in Eqs. (21)–(24).

Using the derived relations in Eqs. (32)–(37) and (41), (42) the limiting procedure may now be applied to the phase-filtered equations, where each quantity has been considered in turn,

$$\lim_{\Delta_f\to0} \frac{\partial}{\partial t}(\theta_k\overline{\rho}_k\tilde{\psi}_k) + \lim_{\Delta_f\to0}\nabla\cdot(\theta_k\overline{\rho}_k\tilde{\psi}_k\tilde{\mathbf{v}}_k) - \lim_{\Delta_f\to0}\nabla\cdot\mathscr{T}_k + \lim_{\Delta_f\to0}\cdot(\theta_k\overline{\rho}_k\tilde{\mathbf{j}}_k) - \lim_{\Delta_f\to0}\theta_k\overline{\rho}_k\tilde{S}_k$$

$$= \lim_{\Delta_f\to0}\left[-\left(\overline{\psi_k\dot{m}_k''} + \overline{\rho_k\mathbf{j}_k\cdot\mathbf{e}_{n,k}^{\mathrm{I}}}\right)_s\oint_{\partial\Omega_k}\mathrm{d}^2x'G(\mathbf{x}'-\mathbf{x})\right] \tag{43}$$

yielding

$$\frac{\partial}{\partial t}(\phi_k\rho_k\psi_k) + \nabla\cdot(\phi_k\rho_k\psi_k\mathbf{v}_k) + \nabla\cdot(\phi_k\rho_k\mathbf{j}_k) - \phi_k\rho_kS_k = -(\psi_k^{\mathrm{I}}\dot{m}_k''^{\mathrm{I}} + \rho_k^{\mathrm{I}}\mathbf{j}_k^{\mathrm{I}}\cdot\mathbf{e}_{n,k}^{\mathrm{I}})a^{\mathrm{I}} \tag{44}$$

Appropriately substituting for the fields ψ_k, $\mathbf{j}_k$, and S_k yields the local instantaneous equations of mass, momentum, species' mass fraction and sensible enthalpy conservation for a separated two-phase flow:

$$\frac{\partial}{\partial t}(\phi_k\rho_k) + \nabla\cdot(\phi_k\rho_k\mathbf{v}_k) = -\dot{m}_k''^{\mathrm{I}}a^{\mathrm{I}} \tag{45}$$

$$\frac{\partial}{\partial t}(\phi_k\rho_k\mathbf{v}_k) + \nabla\cdot(\phi_k\rho_k\mathbf{v}_k\otimes\mathbf{v}_k) - \nabla\cdot(\phi_k\underset{\sim}{\boldsymbol{\sigma}}_k) - \phi_k\rho_k\mathbf{f}_k = -(\mathbf{v}_k^{\mathrm{I}}\dot{m}_k''^{\mathrm{I}} + \underset{\sim}{\boldsymbol{\sigma}}_k^{\mathrm{I}}\cdot\mathbf{e}_{n,k}^{\mathrm{I}})a^{\mathrm{I}} \tag{46}$$

$$\frac{\partial}{\partial t}(\phi_k\rho_kY_{\beta,k}) + \nabla\cdot(\phi_k\rho_kY_{\beta,k}\mathbf{v}_k) - \nabla\cdot(\phi_k\rho_k\mathscr{D}_{m,k}\nabla Y_{\beta,k}) - \phi_k\rho_kS_{\beta,k}$$

$$= -(Y_{\beta,k}^{\mathrm{I}}\dot{m}_k''^{\mathrm{I}} - \rho_k^{\mathrm{I}}\mathscr{D}_{m,k}\nabla Y_{\beta,k}^{\mathrm{I}}\cdot\mathbf{e}_{n,k}^{\mathrm{I}})a^{\mathrm{I}} \tag{47}$$

$$\frac{\partial}{\partial t}(\phi_k\rho_kh_k) + \nabla\cdot(\phi_k\rho_kh_k\mathbf{v}_k) - \nabla\cdot\left(\frac{\phi_k\mu_k}{Sc_k}\nabla h_k\right) - \phi_k\rho_kS_{h,k} = -\left(h_k^{\mathrm{I}}\dot{m}_k''^{\mathrm{I}} - \frac{\mu_k}{Sc_k}\nabla h_k^{\mathrm{I}}\cdot\mathbf{e}_{n,k}^{\mathrm{I}}\right)a^{\mathrm{I}} \tag{48}$$

To summarize this section, the local instantaneous equations for a separated two-phase flow are developed starting from the phase-filtered form without resorting to measure-theoretic statements about the phase interface. Also, a new mathematical interpretation of the local instantaneous interfacial area concentration, a^I is developed and given in Eq. (42), and is shown to be mathematically equivalent to that defined by Kataoka (1986). The local instantaneous separated two-phase flow Eqs. (45)–(48) and (27) will now be used to derive the two-phase FDF transport equation for LES in the next section, and in Section 6.1 the result derived in Eq. (42) will be used when analyzing phase-coupling in the context of the FDF formulation.

5. FMDF method for LES of two-phase turbulent chemically reactive flow

In a fully turbulent single-phase flow, the number of degrees of freedom needed to deterministically describe the system is of the order $Re^{9/4}$ (Pozorski and Minier, 1999). A representative Reynolds number for a flow of interest to the engineer might be $Re \approx 10^5\text{--}10^8$, hence it is clear that one must seek a reduced description of the field for any practical calculation and the *turbulence problem* begins: the number of degrees of freedom in the system must be reduced while the salient physics is retained. Formally reducing the number of degrees of freedom in the system is frequently done by reducing the deterministic field to a statistical description. As a consequence of reducing the number of degrees of freedom, the statistical mean-field is necessarily unclosed. The unclosed nature of the mean-field equations represents the loss of physics contained in the original deterministic equations. The PDF method provides a reduced description of the continuum field that is probabilistic in nature. In the PDF approach, averages of all field variables that are elements of the probability space over which the PDF is defined appear in closed form. Similarly, in FDF methods, phase-filtered values of all field variables that are elements of the phase-space over which the FDF is defined also appear in closed form (Pope, 2000; Gicquel et al., 2002). The advantage of PDF/FDF approaches over other mean-field (i.e., moment-closure) approaches is that for a chemically reactive flow, chemical reaction and radiation/emission source terms appear in closed form (Pope, 1985). Reaction source terms can appear (and usually do) as highly nonlinear functions of temperature and hence representing them in terms of the mean and variance of the temperature (in a 2nd order moment closure, for example) yields a poor approximation at best. The closure issue still exists in the PDF/FDF formalism, however, as terms in the original field equations that have at least one independent variable not contained in the probability, or phase spaces are unclosed and must be modeled in the PDF/FDF transport equations. This situation will always result by virtue of the contracted description; for example, if all field information is represented in the probability space, the PDF equation is exactly the classically deterministic Liouville equation constructed from the original field equations (Pozorski and Minier, 1999; Pathria, 1996).

The starting point of the FDF approach pursued here is the definition of the joint fine-grained density function (JFGDF) for velocity, $\mathbf{u}_k$, and field scalars, $\Psi_{\alpha,k}$, for a two-phase flow (Zhu, 1996). It should be noted here that $\psi_{\alpha,k} = \{Y_{\beta,k}, h_k\}$ should not be confused with the conservation quantity ψ_k of the previous section; $\psi_{\alpha,k}$ represents physical scalar variables only.

$$\xi(\mathbf{u}_1, \mathbf{u}_2, \Psi_{\alpha,1}, \Psi_{\alpha,2}; \mathbf{x}, t) \equiv \phi_1(\mathbf{x}, t) \prod_{i=1}^{3} \delta[v_{1,i}(\mathbf{x}, t) - u_{1,i}] \prod_{\alpha=1}^{\sigma} \delta[\psi_{\alpha,1}(\mathbf{x}, t) - \Psi_{\alpha,1}]$$
$$+ \phi_2(\mathbf{x}, t) \prod_{i=1}^{3} \delta[v_{2,i}(\mathbf{x}, t) - u_{2,i}] \prod_{\alpha=1}^{\sigma} \delta[\psi_{\alpha,2}(\mathbf{x}, t) - \Psi_{\alpha,2}] = \phi_1 \xi_1 + \phi_2 \xi_2 \quad (49)$$

where, $\Psi_{\alpha,k}$ and $\mathbf{u}_k$ are the sampled values of the random variables $\psi_{\alpha,k}$ and $\mathbf{v}_k$ in phase k; at a particular spatial location $\mathbf{x}$ and time t, respectively. On the last line of Eq. (49), the marginal JFGDF for phase 1 is given by ξ_1 and the marginal JFGDF for phase 2 is given by ξ_2. Differentiating Eq. (49) with respect to time yields

$$\frac{\partial \xi}{\partial t} = \left(\phi_1 \frac{\partial \xi_1}{\partial t} + \xi_1 \frac{\partial \phi_1}{\partial t} \right) + \left(\phi_2 \frac{\partial \xi_2}{\partial t} + \xi_2 \frac{\partial \phi_2}{\partial t} \right) \quad (50)$$

Taking the spatial gradient of Eq. (49) yields

$$\nabla \xi = (\phi_1 \nabla \xi_1 + \xi_1 \nabla \phi_1) + (\phi_2 \nabla \xi_2 + \xi_2 \nabla \phi_2) \quad (51)$$

Adding the term $\mathbf{v}_1 \cdot \nabla(\phi_1\xi_1) + \mathbf{v}_2 \cdot \nabla(\phi_2\xi_2)$ to the left and right hand sides of Eq. (50) and using Eq. (51), yields, after some manipulation,

$$
\begin{aligned}
\frac{\partial \xi}{\partial t} + \mathbf{v}_1 \cdot (\phi_1\xi_1) + \mathbf{v}_2 \cdot \nabla(\phi_2\xi_2) = {}& \xi_1\left[\frac{\partial \phi_1}{\partial t} + \mathbf{v}_1 \cdot \nabla\phi_1\right] + \xi_2\left[\frac{\partial \phi_2}{\partial t} + \mathbf{v}_2 \cdot \nabla\phi_2\right] \\
& - \frac{\partial}{\partial \mathbf{u}_1}\left[\phi_1\left(\frac{\partial \mathbf{v}_1}{\partial t} + \mathbf{v}_1 \cdot \nabla\mathbf{v}_1\right)\xi_1\right] - \frac{\partial}{\partial \mathbf{u}_2}\left[\phi_2\left(\frac{\partial \mathbf{v}_2}{\partial t} + \mathbf{v}_2 \cdot \nabla\mathbf{v}_2\right)\xi_2\right] \\
& - \frac{\partial}{\partial \Psi_{\alpha,1}}\left[\phi_1\left(\frac{\partial \psi_{\alpha,1}}{\partial t} + \mathbf{v}_1 \cdot \nabla\psi_{\alpha,1}\right)\xi_1\right] \\
& - \frac{\partial}{\partial \Psi_{\alpha,2}}\left[\phi_2\left(\frac{\partial \psi_{\alpha,2}}{\partial t} + \mathbf{v}_2 \cdot \nabla\psi_{\alpha,2}\right)\xi_2\right]
\end{aligned}
\tag{52}
$$

Collecting terms and summing over both phases, yields

$$
\begin{aligned}
\frac{\partial \xi}{\partial t} + \sum_{k=1}^{2} \mathbf{v}_k \cdot \nabla(\phi_k\xi_k) = {}& \sum_{k=1}^{2} \xi_k\left[\frac{\partial \phi_k}{\partial t} + \mathbf{v}_k \cdot \nabla\phi_k\right] - \sum_{k=1}^{2}\frac{\partial}{\partial \mathbf{u}_k}\left[\phi_k\left(\frac{\partial \mathbf{v}_k}{\partial t} + \mathbf{v}_k \cdot \nabla\mathbf{v}_k\right)\xi_k\right] \\
& - \sum_{k=1}^{2}\frac{\partial}{\partial \Psi_{\alpha,k}}\left[\phi_k\left(\frac{\partial \psi_{\alpha,k}}{\partial t} + \mathbf{v}_k \cdot \nabla\psi_{\alpha,k}\right)\xi_k\right]
\end{aligned}
\tag{53}
$$

Substituting the instantaneous two-phase conservation Eqs. (45)–(48) into Eq. (53) and using the equation of motion for ϕ_k, given by Eq. (27), yields the JFGDF transport equation in the state-space of velocity $\mathbf{u}_k$ and scalars $\Psi_{\alpha,k}$

$$
\begin{aligned}
\frac{\partial \xi}{\partial t} + \mathbf{v}_1 \cdot \nabla(\phi_1\xi_1) + \mathbf{v}_2 \cdot \nabla(\phi_2\xi_2) = {}& \xi_1\Pi_1 + \xi_2\Pi_2 - \frac{\partial}{\partial \mathbf{u}_1}\left[\phi_1\left(-\frac{1}{\rho_1}\nabla(P_1) + \frac{1}{\rho_1}\nabla \cdot (\underset{\sim}{\boldsymbol{\tau}}_1) + \mathbf{f}_1\right)\xi_1\right] \\
& - \frac{\partial}{\partial \mathbf{u}_2}\left[\phi_2\left(-\frac{1}{\rho_2}\nabla(P_2) + \frac{1}{\rho_2}\nabla \cdot (\underset{\sim}{\boldsymbol{\tau}}_2) + \mathbf{f}_2\right)\xi_2\right] \\
& - \frac{\partial}{\partial \Psi_{\alpha,1}}\left[\phi_1\left(-\frac{1}{\rho_1}\nabla \cdot (\mathbf{J}_{\alpha,1}) + S_{\alpha,1}\right)\xi_1\right] \\
& - \frac{\partial}{\partial \Psi_{\alpha,2}}\left[\phi_2\left(-\frac{1}{\rho_2}\nabla \cdot (\mathbf{J}_{\alpha,2}) + S_{\alpha,2}\right)\xi_2\right]
\end{aligned}
\tag{54}
$$

where the Cauchy stress has been decomposed into a thermodynamic pressure P and deviatoric stress $\underset{\sim}{\boldsymbol{\tau}}$ for a Newtonian fluid for each phase.

5.1. The TVSFMDF and conditionally filtered quantities

From the JFGDF, the two-phase velocity-scalar filtered mass density function (TVSFMDF) is defined using the filtering procedure outlined for phase filtering in Section 3 (Jaberi et al., 1999)

$$
F_L(\mathbf{u}_k, \Psi_{\alpha,k}; \mathbf{x}, t) \equiv \langle \rho_k \xi(\mathbf{u}_k, \Psi_{\alpha,k}; \mathbf{x}, t)\rangle = \sum_{k=1}^{2} \int_{\infty} \mathrm{d}^3 x' \rho_k(\mathbf{x}', t) G(\mathbf{x}' - \mathbf{x})\phi_k(\mathbf{x}', t)\xi_k(\mathbf{u}_k, \Psi_{\alpha,k}; \mathbf{x}', t).
\tag{55}
$$

Eq. (55) is the mass weighted spatially filtered fine-grained density. For a general filter G that is everywhere non-negative, i.e., Eq. (7), and satisfies the normalization condition, i.e., Eq. (9), the FMDF has all the properties of a joint probability density function (Pope, 1990, 2000; Givi, 1989). The fundamental integral property of the FMDF is

$$
\int_{\infty} \mathrm{d}^3 u_k d^\sigma \Psi_{\alpha,k}, F_L(\mathbf{u}_k \Psi_{\alpha,k}) = \langle \rho_k(\mathbf{x}, t)\rangle.
\tag{56}
$$

The filtered mass density function, F_L, contains all the one-point statistical information of the marginal FMDF's for both phases. The general relationship between the FMDF and the FDF, f_L, is given by (Jaberi

et al., 1999), $\hat{\rho}(\Psi)f_L(\mathbf{u}, \Phi_\alpha) = F_L(\mathbf{u}, \Psi_\alpha)$, where the meaning of $\hat{Q}(\Psi)$ is given by Eq. (58). Once F_L is known, all one-point statistical information about the two-phase flow may be obtained. Because of this, the TVSFMDF is the primary object of interest in the FDF method and the goal is to determine its time evolution.

Conditionally filtered quantities arise when a filtered quantity contains one or more independent variables that are not an element of the phase-space. The conditional filtered value, $\langle Q_k|_{\chi=\varphi}\rangle$, of a quantity $Q_k(\mathbf{x}, t)$ is defined as

$$\langle Q_k|_{\chi=\varphi}\rangle \equiv \frac{\int \mathrm{d}^3x' G\rho_k\xi Q_k}{\int \mathrm{d}^3x' G\rho_k\xi} = \frac{\int \mathrm{d}^3x' G\rho_k\xi Q_k}{F_L} \tag{57}$$

where $\varphi \equiv \{\mathbf{v}_k, \psi_{\alpha,k}\}$ are the sampled fields associated with the random phase-space fields, $\chi \equiv \{\mathbf{u}_k, \Psi_{\alpha,k}\}$. As mentioned, the conditional filtered Q_k arises when Q_k is not solely a function of φ; accordingly, if Q_k is completely determined by φ then

$$\langle Q_k|_{\chi=\varphi}\rangle = \frac{\int \mathrm{d}^3x' G\rho_k\xi Q_k(\chi)}{\int \mathrm{d}^3x' \rho_k\xi} = \widehat{Q}_k(\varphi). \tag{58}$$

As noted in Section 3, if the field variable $Q_k(\mathbf{x}, t)$ was a chemical reaction source term in Arrhenius form, for example, the conditional filtered value of this quantity would be given directly in closed form assuming the temperature was included in the state-space. This can be seen directly in Eq. (58). In the context of the two-fluid description using the characteristic function, $\gamma(\mathbf{x}, t)$, conditional filtered values may be given by phase (Zhu, 1996)

$$\langle Q|_{\chi=\varphi}\rangle \equiv \langle Q|_{\gamma\geqslant 0^+}\rangle + \langle Q|_{\gamma\leqslant 0^-}\rangle \tag{59}$$

where

$$\langle Q|_{\gamma<0^+}\rangle = \langle Q|_{\chi_1=\varphi_1, \phi_1=1, \chi_2=\phi_2=0}\rangle \tag{60}$$

$$\langle Q|_{\gamma>0^-}\rangle = \langle Q|_{\chi_2=\varphi_2, \phi_2=1, \chi_1=\phi_1=0}\rangle \tag{61}$$

It is clear that only one of the conditional filtered values on the r.h.s. of Eq. (59) may be non-zero at any given time as the characteristic function, $\gamma(\mathbf{x}, t)$, is a single valued function of $\mathbf{x}$ and t. That is, γ specifies *either* phase 1, phase 2, *or* the phase interface at a given spatial location $\mathbf{x}$ at time t. Furthermore, the conditionally filtered gradient of field scalar $\psi_k(\mathbf{x}, t)$ can be decomposed into a component within the phase and on the phase interface by a simple application of the chain rule of differential calculus and the knowledge that ϕ_k is constant within either phase. Considering phase k,

$$\langle \phi_k\nabla\psi_k|_{\gamma\leqslant 0^-}\rangle = \langle \nabla(\phi_k\psi_k)|_{\gamma\leqslant 0^-}\rangle - \langle \psi_k\nabla\phi_k|_{\gamma=0^-}\rangle \tag{62}$$

It can be shown that if the volumetric interphase conversion term, Π_k, is assumed to be negligible (see Eq. (88)), the conditional average in the first term on the r.h.s. of Eq. (62) is valid within the phase and Eq. (62) becomes

$$\langle \phi_k\nabla\psi_k|_{\gamma\leqslant 0^-}\rangle = \langle \nabla(\phi_k\psi_k)|_{\gamma<0^-}\rangle - \langle \psi_k\nabla\phi_k|_{\gamma=0^-}\rangle \tag{63}$$

It is advantageous to cast the phase-coupling terms in the form of Eq. (62) as using Eqs. (40) and (41), it follows that the last term on the r.h.s. of Eq. (62) may be expressed as

$$\langle \psi_k\nabla\phi_k|_{\gamma=0^-}\rangle = \langle \psi_k\delta(\gamma)\nabla\gamma|_{\gamma=0^-}\rangle = \left\langle \psi_k|\nabla_\gamma|\delta(\gamma)\left(\frac{\nabla\gamma}{|\nabla\gamma|}\right)\bigg|_{\gamma=0^-}\right\rangle = \left\langle \psi_k^{\mathrm{I}}a^{\mathrm{I}}\mathbf{e}_{n,k}^{\mathrm{I}}\bigg|_{\gamma=0^-}\right\rangle. \tag{64}$$

Thus, it is seen that the contribution on the phase interface may be expressed in terms of the interfacial area concentration, a^{I}, consistent with the phase-coupling source terms found in Section 4. In the TVSFMDF transport equation, conditionally surface-filtered terms like Eq. (64) characterize phase coupling and are of central importance in the formulation, as will be discussed in Section 6.

The anticipated application of this two-phase FMDF formulation is in the simulation of a fully coupled dilutely dispersed phase-continuum phase flow; viz. a dilute droplet laden or particle laden turbulent gas flow. The physical consequences of the dilute flow assumption are covered in more detail in Section 6 of this paper.

5.2. Unclosed FMDF transport equation

Filtering Eq. (54) yields the unclosed TVSFMDF transport equation for a separated two-phase flow, and may be written as follows:

$$
\frac{\partial F_L}{\partial t} + \frac{\partial}{\partial \mathbf{x}}\left[\langle \phi_1 \mathbf{v}_1|_{\gamma \leqslant 0^-}\rangle F_L^1\right] + \frac{\partial}{\partial \mathbf{x}}\left[\langle \phi_2 \mathbf{v}_2|_{\gamma \geqslant 0^+}\rangle F_L^2\right]
$$

$$
= -\frac{\partial}{\partial \mathbf{u}_1}\left[\left\langle \phi_1\left(-\frac{1}{\rho_1}\nabla P_1 + \frac{1}{\rho_1}\nabla \cdot \underset{\sim}{\boldsymbol{\tau}}_1 + \mathbf{f}_1\right)\Big|_{\gamma \leqslant 0^-}\right\rangle F_L^1\right]
$$

$$
- \frac{\partial}{\partial \mathbf{u}_2}\left[\left\langle \phi_2\left(-\frac{1}{\rho_2}\nabla P_2 + \frac{1}{\rho_2}\nabla \cdot \underset{\sim}{\boldsymbol{\tau}}_2 + \mathbf{f}_2\right)\Big|_{\gamma \geqslant 0^+}\right\rangle F_L^2\right]
$$

$$
- \frac{\partial}{\partial \Psi_{\alpha,1}}\left[\left\langle \phi_1\left(-\frac{1}{\rho_1}\nabla \cdot \mathbf{J}_{\alpha,1} + S_{\alpha,1}\right)\Big|_{\gamma \leqslant 0^-}\right\rangle F_L^1\right]
$$

$$
- \frac{\partial}{\partial \Psi_{\alpha,2}}\left[\left\langle \phi_2\left(-\frac{1}{\rho_2}\nabla \cdot \mathbf{J}_{\alpha,2} + S_{\alpha,2}\right)\Big|_{\gamma \geqslant 0^+}\right\rangle F_L^2\right]
$$

$$
+ \langle \Pi_1|_{\gamma = 0^-}\rangle F_L^1 + \langle \Pi_2|_{\gamma = 0^+}\rangle F_L^2 \tag{65}
$$

The interphase conversion terms, Π_k are included here in the general formulation as no approximation regarding the interphase conversion is yet taken; the conditional averages are not separated by phase. It should be noted that in writing Eq. (65), a low Mach number approximation is assumed which results in a de-coupling of pressure and density. The Favre averaged marginal (in phase k) velocity-scalar FMDF is related the TVSFMDF via $\theta_k\langle \rho_k\rangle \tilde{f}_k = \hat{\rho}_k \phi_k f_L$, and $\hat{\rho}_k \phi_k f_L = \hat{\rho}_k f_L^k = F_L^k$. One may multiply Eq. (65) by the phase indicator function, ϕ_k, to extract the marginal FMDF. Because the interface function, $\gamma(\mathbf{x},t)$, is not an element of the phase-space, an additional interphase conversion term, Π_k, results from the commutation of ϕ_k inside the derivatives $\partial/\partial t$ and $\partial/\partial \mathbf{x}$. The result is the marginal velocity-scalar FMDF transport equation written for phase 1:

$$
\frac{\partial F_L^1}{\partial t} + \frac{\partial}{\partial \mathbf{x}}\left[\left\langle \phi_1 \mathbf{v}_1|_{\gamma \leqslant 0^-}\right\rangle F_L^1\right] = -\frac{\partial}{\partial \mathbf{u}_1}\left[\left\langle \phi_1 \cdot \left(-\frac{1}{\rho_1}\nabla P_1 + \frac{1}{\rho_1}\nabla \cdot \underset{\sim}{\boldsymbol{\tau}}_1 + \mathbf{f}_1\right)\Big|_{\gamma \leqslant 0^-}\right\rangle F_L^1\right]
$$

$$
- \frac{\partial}{\partial \Psi_{\alpha,1}}\left[\left\langle \phi_1\left(-\frac{1}{\rho_1}\nabla \cdot \mathbf{J}_{\alpha,1} + S_{\alpha,1}\right)\Big|_{\gamma \leqslant 0^-}\right\rangle F_L^1\right] + \langle \Pi_1|_{\gamma = 0^-}\rangle F_L^1
$$

$$
+ \left\langle (\mathbf{v}_1 - \mathbf{v}^{\mathrm{I}}) \cdot \mathbf{e}_{n,1}^{\mathrm{I}}|\nabla \gamma|\Big|_{\gamma = 0^-}\right\rangle F_L^1 \tag{66}
$$

As can be seen in Eq. (66), transport of F_L^1 is generated in configuration, velocity and scalar phase-spaces. These phenomenon will be considered in turn starting with those processes that are treated exactly in the velocity-scalar FMDF context: transport of F_L^1 in configuration phase-space and transport in scalar phase-space due to reaction source terms. Following this discussion, divergence and gradient terms in Eq. (66) admit similar decompositions involving evolution of the FMDF due to molecular processes and will therefore be considered together.

It should be noted here that phase-space information may be integrated out of the velocity-scalar FMDF in phase k to extract the scalar-only FMDF and its subsequent transport equation from Eq. (66), according to the relations (Pope, 1985):

$$\int \mathrm{d}^3 u_1 F_L^1(\mathbf{u}_1, \Psi_{\alpha,1}) = F_{L,\Psi}^1(\Psi_{\alpha,1}) \tag{67}$$

$$\int \mathrm{d}^3 u_k \frac{\partial}{\partial \mathbf{u}_k}[\bullet] = 0 \tag{68}$$

In the velocity-scalar FMDF formulation, the phase-space measure in phase 1 includes the velocity in that phase, therefore, the spatial gradient of conditionally averaged velocity is in closed form according to Eq. (58). Hence, the contribution to the transport of the marginal TVSFMDF in configuration phase-space is written as

$$\frac{\partial}{\partial \mathbf{x}}\left[\langle \phi_1 \mathbf{v}_1|_{\gamma \leqslant 0^-}\rangle F_L^1\right] = \frac{\partial}{\partial \mathbf{x}}(\theta_1 \mathbf{u}_1 F_L^1). \tag{69}$$

Thus, we see that convection is treated exactly in the velocity-scalar FMDF context. Similarly, the change in F_L^1 in scalar phase-space due to source terms in Eq. (66) is given in closed from if the scalar source terms S_α are completely specified in terms of scalar phase-space variables; i.e., included in the phase 1 phase-space measure. Therefore, if $S_\alpha = f(\Psi_\alpha, \mathbf{u})$ then, according to Eq. (58),

$$\frac{\partial}{\partial \Psi_{\alpha,1}}\left[\langle \phi_1 S_{\alpha,1}|_{\gamma \leqslant 0^-}\rangle F_L^1\right] = \frac{\partial}{\partial \Psi_{\alpha,1}}(\theta_1 \hat{S}_{\alpha,1} F_L^1). \tag{70}$$

Transport of F_L^1 in velocity phase-space is shown in the first term on the r.h.s. of Eq. (66), and may be written as

$$\frac{\partial}{\partial \mathbf{u}_1}\left[\left\langle \phi_1\left(-\frac{1}{\rho_1}\nabla P_1 + \frac{1}{\rho_1}\nabla \cdot \underset{\sim}{\tau}_1 + \mathbf{f}_1\right)\bigg|_{\gamma \leqslant 0^-}\right\rangle F_L^1\right]$$

$$= -\frac{\partial}{\partial \mathbf{u}_1}\left[\left\langle \frac{1}{\rho_1}\phi_1 \nabla P_1\bigg|_{\gamma \leqslant 0^-}\right\rangle F_L^1\right] + \frac{\partial}{\partial \mathbf{u}_1}\left[\left\langle \frac{1}{\rho_1}\phi_1 \nabla \cdot \underset{\sim}{\tau}_1\bigg|_{\gamma \leqslant 0^-}\right\rangle F_L^1\right] + \frac{\partial}{\partial \mathbf{u}_1}\left[\langle \phi_1 \mathbf{f}_1|_{\gamma \leqslant 0^-}\rangle F_L^1\right] \tag{71}$$

where, according to Eqs. (63) and (64), the components including pressure and deviatoric stress are

$$\frac{\partial}{\partial \mathbf{u}_1}\left[\left\langle \frac{1}{\rho_1}\phi_1 \nabla P_1\bigg|_{\gamma \leqslant 0^-}\right\rangle F_L^1\right] = \frac{\partial}{\partial \mathbf{u}_1}\left[\left\langle \frac{1}{\rho_1}\nabla(\phi_1 P_1)\bigg|_{\gamma < 0^-}\right\rangle F_L^1\right] - \frac{\partial}{\partial \mathbf{u}_1}\left[\left\langle \frac{1}{\rho_1}P_1^\mathrm{I} a^\mathrm{I} \mathbf{e}_{n,1}^\mathrm{I}\bigg|_{\gamma = 0^-}\right\rangle F_L^1\right] \tag{72}$$

and,

$$\frac{\partial}{\partial \mathbf{u}_1}\left[\left\langle \frac{1}{\rho_1}\phi_1 \nabla \cdot \underset{\sim}{\tau}_1\bigg|_{\gamma \leqslant 0^-}\right\rangle F_L^1\right] = \frac{\partial}{\partial \mathbf{u}_1}\left[\left\langle \frac{1}{\rho_1}\nabla \cdot (\phi_1 \underset{\sim}{\tau}_1)\bigg|_{\gamma < 0^-}\right\rangle F_L^1\right] - \frac{\partial}{\partial \mathbf{u}_1}\left[\left\langle \frac{1}{\rho_1}\underset{\sim}{\tau}_1^\mathrm{I} a^\mathrm{I} \cdot \mathbf{e}_{n,1}^\mathrm{I}\bigg|_{\gamma = 0^-}\right\rangle F_L^1\right] \tag{73}$$

The first term on the r.h.s. of Eq. (73) represents the conditional diffusion of momentum and may be decomposed into a conditional dissipation term (Pope, 1985) as follows,

$$-\frac{\partial}{\partial \mathbf{u}_1}\left[\left\langle \frac{1}{\rho_1}\nabla \cdot \left(\phi_1 \underset{\sim}{\tau}_1\right)\bigg|_{\gamma < 0^-}\right\rangle F_L^1\right] = -\frac{\partial}{\partial \mathbf{u}_1}\left[\left\langle \frac{\mu_1}{\rho_1}\nabla^2(\phi_1 \mathbf{v}_1)\bigg|_{\gamma < 0^-}\right\rangle F_L^1\right]$$

$$= \theta_1 \mu_1 \nabla^2(F_L^1/\hat{\rho}_1) - \frac{\partial}{\partial \mathbf{u}_1}\left[\frac{\partial}{\partial \mathbf{u}_1}\left\langle \phi_1 \mu_1 \nabla \mathbf{v}_1 \cdot \nabla \mathbf{v}_1|_{\gamma < 0^-}\right\rangle F_L^1/\widehat{\rho_1}\right]$$

$$- \frac{\partial}{\partial \mathbf{u}_1}\left[\frac{\partial}{\partial \Psi_{\alpha,1}}\left\langle \phi_1 \mu_1 \nabla \mathbf{v}_1 \cdot \nabla \Psi_{\alpha,1}|_{\gamma < 0^-}\right\rangle F_L^1/\widehat{\rho_1}\right] \tag{74}$$

The first term on the r.h.s. of Eq. (74) after the Laplacian of the marginal TVSFMDF is the conditional dissipation in the momentum phase-space and physically represents the effect of SGS mixing of the momentum on the transport of F_L^1. The second term on the r.h.s. is conditional dissipation in momentum and scalar phase-space and represents the diffusion of F_L^1 due to the combination of scalar and momentum fluxes. Using a

similar conditional dissipation decomposition as used in Eq. (74), the transport of F_L^1 in scalar phase-space due to the gradient flux $\mathbf{J}_{\alpha,1}$ is written as

$$
\begin{aligned}
\frac{\partial}{\partial \Psi_{\alpha,1}} \left[\left\langle \frac{1}{\rho_1} \phi_1 \nabla \cdot \mathbf{J}_{\alpha,1} \Big|_{\gamma \leqslant 0^-} \right\rangle F_L^1 \right] &= -\frac{\partial}{\partial \Psi_{\alpha,1}} \left[\left\langle \frac{1}{\rho_1} \nabla \cdot (\phi_1 \mathbf{J}_{\alpha,1}) \Big|_{\gamma < 0^-} \right\rangle F_L^1 \right] \\
&\quad + \frac{\partial}{\partial \Psi_{\alpha,1}} \left[\left\langle \frac{1}{\rho_1} \mathbf{J}_{\alpha,1}^{\mathrm{I}} \cdot \mathbf{e}_{n,1}^{\mathrm{I}} a^{\mathrm{I}} \Big|_{\gamma = 0^-} \right\rangle F_L^1 \right] \\
&= \frac{\partial}{\partial \Psi_{\alpha,1}} \left[\left\langle \frac{1}{\rho_1} \nabla \cdot (\phi_1 \Gamma_{\alpha,1} \nabla \psi_{\alpha,1}) \Big|_{\gamma < 0^-} \right\rangle F_L^1 \right] \\
&\quad + \frac{\partial}{\partial \Psi_{\alpha,1}} \left[\left\langle \frac{1}{\rho_1} \Gamma_{\alpha,1} \nabla \psi_{\alpha,1}^{\mathrm{I}} \cdot \mathbf{e}_{n,1}^{\mathrm{I}} a^{\mathrm{I}} \Big|_{\gamma = 0^-} \right\rangle F_L^1 \right] = -\theta_1 \Gamma_{\alpha,1} \nabla^2 (F_L^1 / \widehat{\rho_1}) \\
&\quad + \frac{\partial}{\partial \Psi_{\alpha,1} \partial \Psi_{\beta,1}} \left[\left\langle \phi_1 \Gamma_{\alpha,1} \nabla \psi_{\alpha,1} \cdot \nabla \psi_{\beta,1} \big|_{\gamma < 0^-} \right\rangle (F_L^1 / \widehat{\rho_1}) \right] \\
&\quad + \frac{\partial}{\partial \Psi_{\alpha,1} \partial \mathbf{u}_1} \cdot [\langle \phi_1 \Gamma_{\alpha,1} \nabla \psi_{\alpha,1} \cdot \nabla \mathbf{v}_1 |_{\gamma < 0^-} \rangle (F_L^1 / \widehat{\rho_1})] \\
&\quad + \frac{\partial}{\partial \Psi_{\alpha,1}} \left[\left\langle \frac{1}{\rho_1} \Gamma_{\alpha,1} \nabla \psi_{\alpha,1}^{\mathrm{I}} \cdot \mathbf{e}_{n,1}^{\mathrm{I}} a^{\mathrm{I}} \Big|_{\gamma = 0^-} \right\rangle F_L^1 \right]
\end{aligned}
\tag{75}
$$

The conditional dissipation in the scalar phase-space, shown in the second term on the r.h.s. of the last equality in Eq. (75), physically represents the effects of SGS mixing of the scalar quantity ψ_α on the transport of F_L^1. The third term on the r.h.s. of Eq. (75) is another conditional dissipation term in the momentum and scalar phase-space, and physically represents the molecular diffusion of F_L^1 due to scalar and momentum flux.

It is important to note that when describing both the momentum and scalar fields in the TVSFMDF, the scalar and velocity state-spaces are coupled as can be seen by the mixed derivatives of $\mathbf{u}_1$ and $\Psi_{\alpha,1}$ in Eqs. (74) and (75). This coupling has a very important consequence when the TVSFMDF is solved. When the Monte-Carlo method is employed to solve the marginal TVSFMDF transport equation, mixed derivatives in state-space variables result in stochastic coupling between independent diffusion processes (in the context of Itô calculus) within the stochastic differential equations used to solve Eq. (66) (Gardiner, 1982; Sheikhi et al., 2003).

Collecting Eqs. (69)–(75), the unclosed marginal TVSFMDF transport equation in phase 1 for a separated two-phase flow, including phase-coupling terms, is written as

$$
\begin{aligned}
\frac{\partial F_L^1}{\partial t} + \frac{\partial}{\partial \mathbf{x}} (\theta_1 \mathbf{u}_1 F_L^1) &= \frac{\partial}{\partial \mathbf{x}} \left[\mu_1 \theta_1 \frac{\partial (F_L^1 / \widehat{\rho_1})}{\partial \mathbf{x}} \right] - \frac{\partial^2}{\partial \mathbf{u}_1 \cdot \partial \mathbf{u}_1} [\langle \mu_1 \phi_1 \nabla \mathbf{v}_1 \cdot \nabla \mathbf{v}_1 |_{\gamma < 0^-} \rangle (F_L^1 / \widehat{\rho_1})] \\
&\quad + \frac{\partial}{\partial \mathbf{x}} \left[\theta_1 \Gamma_{\alpha,1} \frac{\partial (F_L^1 / \widehat{\rho_1})}{\partial \mathbf{x}} \right] - \frac{\partial^2}{\partial \mathbf{u}_1 \partial \Psi_{\alpha,1}} [\langle \mu_1 \phi_1 \nabla \mathbf{v}_1 \cdot \nabla \psi_{\alpha,1} |_{\gamma < 0^-} \rangle (F_L^1 / \widehat{\rho_1})] \\
&\quad - \frac{\partial}{\partial \Psi_{\alpha,1}} [\theta_1 S_{\alpha,1} F_L^1] - \frac{\partial^2}{\partial \Psi_{\alpha,1} \partial \Psi_{\beta,1}} [\langle \phi_1 \Gamma_{\alpha,1} \nabla \psi_{\alpha,1} \cdot \nabla \psi_{\beta,1} |_{\gamma < 0^-} \rangle (F_L^1 / \widehat{\rho_1})] \\
&\quad + \frac{\partial}{\partial \mathbf{u}_1} \left[\left\langle \frac{1}{\rho_1} \nabla (\phi_1 P_1) \Big|_{\gamma < 0^-} \right\rangle F_L^1 \right] - \frac{\partial^2}{\partial \Psi_{\alpha,1} \partial \mathbf{u}_1} [\langle \phi_1 \Gamma_{\alpha,1} \nabla \psi_{\alpha,1} \cdot \nabla \mathbf{v}_1 |_{\gamma < 0^-} \rangle (F_L^1 / \widehat{\rho_1})] \\
&\quad + \frac{\partial}{\partial \mathbf{u}_1} [\langle \phi_1 \mathbf{f}_1 |_{\gamma < 0^-} \rangle F_L^1] + \frac{\partial}{\partial \Psi_{\alpha,1}} \left[\left\langle \frac{1}{\rho_1} \Gamma_{\alpha,1} \nabla \psi_{\alpha,1}^{\mathrm{I}} \cdot \mathbf{e}_{n,1}^{\mathrm{I}} a^{\mathrm{I}} \Big|_{\gamma = 0^-} \right\rangle F_L^1 \right] \\
&\quad - \frac{\partial}{\partial \mathbf{u}_1} \left[\left\langle \frac{1}{\rho_1} P_1^{\mathrm{I}} \mathbf{e}_{n,1}^{\mathrm{I}} a^{\mathrm{I}} \Big|_{\gamma = 0^-} \right\rangle F_L^1 \right] + \frac{\partial}{\partial \mathbf{u}_1} \left[\left\langle \frac{1}{\rho_1} \underset{\sim}{\boldsymbol{\tau}}_1^{\mathrm{I}} \cdot \mathbf{e}_{n,1}^{\mathrm{I}} a^{\mathrm{I}} \Big|_{\gamma = 0^-} \right\rangle F_L^1 \right].
\end{aligned}
\tag{76}
$$

Eq. (76) governs the time evolution of the marginal FMDF, F_L^1 in phase 1 of a two-phase flow. Eq. (76) can be further simplified by integrating the velocity information contained in phase 1 out of Eq. (76) according to

Eqs. (67) and (68) yielding a two-phase scalar-only FMDF transport equation. The resulting scalar FMDF transport equation acquiesces directly with a hybrid LES-FMDF formulation when simpler approaches are pursued to model the momentum field (e.g., eddy viscosity models). In general, all conditionally filtered terms, including those conditioned on the phase interface, are unclosed and must be modeled to close the equation. Specifying closure models here is beyond the scope of this work, however, well known closures exist for the conditional dissipation, conditionally filtered pressure gradient and body force terms for single-phase flows (Pope, 1985; Pandya and Mashayek, 2001; O'Brien, 1980; Fox, 2003; Pozorski and Minier, 1999; Colucci et al., 1998; Zhu, 1996). As a first step, these closures could be applied directly to all terms in Eq. (76) conditionally filtered within the fluid phase, $\gamma < 0^-$. What is unique to the two-phase formulation, however, is the conditionally surface-filtered terms, shown in Eq. (76) conditioned upon $\gamma = 0^-$ (the last three terms in Eq. (76)). These unclosed conditionally surface-filtered terms account for phase interaction and are therefore crucially important in the two-phase formulation. Analysis of these terms is the focus of the following section.

6. Phase-coupling terms in the TVSFMDF

Phase-coupling is arguably the most important part of simulating a multiphase flow. Dealing with phase-coupling terms is difficult, however, because frequently these terms are not well defined. The formulation of the local instantaneous equations in terms of the interfacial area concentration, a^I is a perfect example; a^I is, in fact, a distribution seen directly in Eq. (41), and coupling terms involving a^I must be modeled (Zhu et al., 2000; Rumberg and Rogg, 2000). Multiphase flow modeling with PDF/FDF methods are relatively new and have not received much attention to date compared to single-phase PDF/FDF methods. Furthermore, phase-coupling terms arising in multiphase PDF/FDF formulations have received even less attention due to the difficulties arising in specifying their functional form in addition to added difficulties with respect to solving the transport of the PDF/FDF using Monte-Carlo methods, as will be discussed below.

6.1. Conditionally surface-filtered quantities

Conditionally surface-filtered quantities (i.e., conditioned upon the interface: $\gamma = 0^-$), shown on the r.h.s. of Eq. (76) embody coupling between phase 1 and phase 2 in the two-phase flow. This can be seen directly by considering jump conditions across the interface. Eqs. (45)–(48) represent the conservation equations in each phase k of the two-phase flow, including the phase interface. Summing Eqs. (45)–(48) over both phases produces a total field representation of the conservation equations valid in both phases and the phase interface. Within either phase, the conservation equations hold and $\phi(\mathbf{x}, t) = \text{const}$, however, on the phase interface the conservation equations hold with $\phi(\mathbf{x}, t) \neq \text{const}$, hence the mass-flux, momentum-flux, species' mass fraction-flux and sensible enthalpy-flux local instantaneous jump conditions are obtained from Eqs. (45)–(48) and are written as

$$\sum_{k=1}^{2} \left\{ \rho_k^I (\mathbf{v}_k - \mathbf{v}^I) \cdot \mathbf{e}_{n,k}^I a^I \right\} = 0 \tag{77}$$

$$\sum_{k=1}^{2} \left\{ \rho_k^I \mathbf{v}_k (\mathbf{v}_k - \mathbf{v}^I) \cdot \mathbf{e}_{n,k}^I a^I - (P_k^I \mathbf{e}_{n,k}^I - \underset{\sim}{\tau}_k^I \cdot \mathbf{e}_{n,k}^I) a^I \right\} = \sum_{k=1}^{2} \rho_k^I \boldsymbol{\sigma}_{s,k} a^I \tag{78}$$

$$\sum_{k=1}^{2} \left\{ \rho_k^I Y_{\beta,k}^I (\mathbf{v}_k - \mathbf{v}^I) \cdot \mathbf{e}_{n,k}^I a^I - \rho_k^I \mathscr{D}_{m,k} \nabla Y_{\beta,k}^I \cdot \mathbf{e}_{n,k}^I a^I \right\} = \sum_{k=1}^{2} \rho_k^I S_{\beta,k}''^I a^I \tag{79}$$

$$\sum_{k=1}^{2} \left\{ \rho_k^I h_k^I (\mathbf{v}_k - \mathbf{v}^I) \cdot \mathbf{e}_{n,k}^I a^I - \frac{\mu k}{Sc_k} \nabla h_k^I \cdot \mathbf{e}_{n,k}^I a^I \right\} = \sum_{k=1}^{2} \rho_k^I S_{h,k}''^I a^I \tag{80}$$

where the terms $S_\beta''^I$ and $\boldsymbol{\sigma}_s$ are the surface reaction and surface tension source terms per unit interfacial area, respectively. The term $S_h''^I$ contains the surface energy source terms as well as pressure work and viscous stress work per unit interfacial area. Surface source terms were neglected in the derivation of Eqs. (45)–(48) by virtue

of the form of the phase-filtered equations in Section 3 and are included here for completeness. The interested reader is referred to Kuo (1986) and Kataoka (1986) for details.

The instantaneous jump conditions shown in Eqs. (77)–(80) allow one to relate fluxing quantities in one phase to those in the other, however, these jump conditions do not shed any light on the closure issue involving conditionally surface-filtered terms in Eq. (76). Recall from Eq. (57), the conditional filter of an interface quantity $1/\rho_1 Q_1(\mathbf{x}, t) a^{\mathrm{I}}$, conditioned on the surface $\gamma = 0^-$ is given by

$$\left\langle \frac{1}{\rho_1} Q_1 a^{\mathrm{I}} \bigg|_{\gamma=0^-} \right\rangle = \frac{\int_{\Omega_{\mathrm{T}}} \mathrm{d}^3 x' G \xi Q_1 a^{\mathrm{I}}}{\int_{\Omega_{\mathrm{T}}} \mathrm{d}^3 x' G \rho_1 \xi} \tag{81}$$

Recall also, the instantaneous area concentration shown in Eq. (42), and rewritten below, may be expressed in terms of a surface integration of the Dirac distribution,

$$a^{\mathrm{I}} = \oint_{\partial \Omega_1} \mathrm{d}^2 x' \delta(\mathbf{x}' - \mathbf{x}) \tag{82}$$

Replacing a^{I} in the numerator integral of Eq. (81) with Eq. (82) and by mollifying the phase velocity (Evans, 2000; Pitman, 2005) and requiring $\Psi_{\alpha,1}$ to have finite mean and variance, Fubini's theorem may be applied to write (Choquet-Bruhat et al., 1982)

$$\begin{aligned}
\int_{\Omega_{\mathrm{T}}} \mathrm{d}^3 x' G \xi Q_1 a^{\mathrm{I}} &= \int_{\Omega_{\mathrm{T}}} \mathrm{d}^3 x' \left\{ G \xi Q_1 \oint_{\partial \Omega_1} \mathrm{d}^3 x'' \delta(\mathbf{x}'' - \mathbf{x}') \right\} \\
&= \oint_{\partial \Omega_1} \mathrm{d}^2 x'' \left\{ \int_{\Omega_{\mathrm{T}}} \mathrm{d}^3 x' G(\mathbf{x}' - \mathbf{x}) Q_1(\mathbf{x}', t) \xi(\mathbf{x}', t) \delta(\mathbf{x}'' - \mathbf{x}') \right\} \\
&= \oint_{\partial \Omega_1} \mathrm{d}^2 x'' G(\mathbf{x}'' - \mathbf{x}) Q_1(\mathbf{x}'', t) \xi(\mathbf{x}'', t) \tag{83}
\end{aligned}$$

Therefore the volume integration can be related to a surface integration on the phase interface due to the local instantaneous area concentration, a^{I}. Consider that because the volume fraction θ_1 in phase 1 is given as the filtered value of the local instantaneous volume fraction ϕ_1 in that phase (i.e., $\theta_1 = \langle \phi_1 \rangle$) and recalling that the FGDF for a separated two-phase flow is given by $\xi = \phi_1 \xi_1 + \phi_2 \xi_2$ according to Eq. (49) with $\phi_2 \equiv 0$ in phase 1 (see Eq. (26)), Eq. (18) may be used to express the denominator of Eq. (81) as follows:

$$\int_{\Omega_{\mathrm{T}}} \mathrm{d}^3 x' G \rho_1 \xi = \frac{V_1}{A_1} \oint_{\partial \Omega_1} \mathrm{d}^2 x' G \rho_1 \xi_1 \tag{84}$$

where A_1 and V_1 are the area and volume occupied by phase 1 at time t. In Eqs. (83) and (84), the FGDF selects only those values of Q_1 and ρ_1 that lie on the phase interface as the constraint $\gamma = 0 \Rightarrow \mathbf{x} = \mathbf{x}^{\mathrm{I}}$ must hold. The result is that all terms in Eqs. (83) and (84) are integrated on the phase-boundary by virtue of $\gamma(\mathbf{x})$ and only those Q_1 and ρ_1 that live on the surface are kept by virtue of ξ (Kuo, 1986). Using the result in Eqs. (83) and (84) to re-cast Eq. (81) for a *surface-filtered* quantity $1/\rho_1 Q_1(\mathbf{x}, t) a^{\mathrm{I}}$, the result is

$$\left\langle \frac{1}{\rho_1} Q_1 a^{\mathrm{I}} \bigg|_{\gamma=0^-} \right\rangle = \frac{\oint_{\partial \Omega_1} \mathrm{d}^2 x' G Q_1^{\mathrm{I}}}{\oint_{\partial \Omega_1} \mathrm{d}^2 x' G \rho_1^{\mathrm{I}}} \tag{85}$$

Because the surface integrals in Eq. (85) are taken on the phase interface, the superfluous superscript I notation inside the integrands can be disregarded. From Eq. (17) the above can now be re-written in terms of surface-filtered quantities

$$\left\langle \frac{1}{\rho_1} Q_1 a^{\mathrm{I}} \bigg|_{\gamma=0^-} \right\rangle = \frac{A_1}{V_1} \frac{\overline{(Q_1)_s}}{\overline{(\rho_1)_s}}. \tag{86}$$

Eq. (86) reveals a *direct link* between the conditionally filtered surface exchange terms in Eq. (76) and the PCT's in Eqs. (21)–(24). The benefit of establishing this connection is that existing models for the PCT's may be used directly in the TVSFMDF transport equation.

6.2. Conditionally filtered interphase volume conversion

The last two terms on the r.h.s. of Eq. (66) represent the average volumetric rate of change of phase 2 due to interphase conversion with phase 1. Geometrically, as seen in Eq. (27), $\Pi_k(\mathbf{x}, t)$ balances the convective and pure-transient rate of change of a point on the phase interface at a position $\mathbf{x}^I$ and time t. Physically, $\Pi_1(\mathbf{x}, t)$ tracks how the size of phase 1 changes in response to change in size of phase 2 due to momentum transfer between phases, when compressibility of one or both phases is important, and mass and/or energy transfer between phases. The functional form of Π_1 can be more clearly stated by using the definition of the phase indicator function shown in Eq. (26) and applying the kinematic boundary condition at the phase interface (Dean and Dalrymple, 1995), in light of Eqs. (41) and (42), the interphase conversion term, Π_k, is specified as

$$\frac{\partial \phi_k}{\partial t} + \mathbf{v}_k \cdot \nabla \phi_k = \Pi_k = -\mathbf{v}^I \cdot \nabla \phi_k + \mathbf{v}_k \cdot \nabla \phi_k = \delta(\gamma) \nabla \gamma \cdot (\mathbf{v}_k - \mathbf{v}^I) = (-1)^k a^I (\mathbf{v}_k - \mathbf{v}^I) \cdot \mathbf{e}_{n,k}^I. \tag{87}$$

Comparing Eq. (87) to Eq. (77), it is clear that $\Pi_k = (-1)^k (\mathbf{v}_k - \mathbf{v}^I) \cdot \mathbf{e}_{n,k}^I a^I$ is determined by the mass flux jump condition across the phase interface

$$\sum_{k=1}^{2} \rho_k^I (\mathbf{v}_k - \mathbf{v}^I) \cdot \mathbf{e}_{n,k}^I a^I = \sum_{k=1}^{2} \rho_k \Pi_k = 0. \tag{88}$$

Previous work has attributed $\Pi_k(\mathbf{x}, t)$ to coalescence and break-up for a discrete dispersed liquid droplet phase, and subsequently disregarded when the dispersed phase is dilute (Zhu, 1996; Drew and Passman, 1998). Specification of this term and conditions under which it may be disregarded frequently leads to confusion in the multiphase flow literature. Following directly the development of Drew and Passman (1998) for a dispersed droplet or bubble phase, let $f(V, \mathbf{x}, t)$ be the number density of spherical droplets or bubbles of volume V. The probability of finding a droplet with volume between V and $V + \mathrm{d}V$ within a $\mathrm{d}\mathbf{x}$ of $\mathbf{x}$ is given by $f(V, \mathbf{x}, t)\mathrm{d}V\mathrm{d}^3x$. An evolution equation for f is *assumed* as

$$\frac{\partial f}{\partial t} + \nabla \cdot (f\mathbf{v}_d) = \left.\frac{\mathrm{d}f}{\mathrm{d}t}\right|_c + \left.\frac{\mathrm{d}f}{\mathrm{d}t}\right|_b \tag{89}$$

where $\mathbf{v}_d$ is the velocity associated with droplets of volume V and the total derivatives with subscripts on the r.h.s. of Eq. (89) correspond to the rate of change of the *number* of droplets due to coalescence and breakup, respectively. The number density, identifying phase 1 with the dispersed droplets, is then given by

$$n_1(\mathbf{x}, t) = \int_{\Omega_T} \mathrm{d}V f(V, \mathbf{x}, t) \tag{90}$$

The average droplet volume is given in terms of the number density

$$\langle V_1 \rangle = \int_{\Omega_T} \mathrm{d}V \frac{f(V, \mathbf{x}, t)}{n_1(\mathbf{x}, t)} V \tag{91}$$

and the volume fraction is

$$\theta_1(\mathbf{x}, t) = \int_{\Omega_T} \mathrm{d}V\, V_1 f(V, \mathbf{x}, t) = \langle V_1 \rangle n_1(\mathbf{x}, t) \tag{92}$$

It is generally assumed that if two droplets coalesce, the resulting droplet volume is the sum of these droplets and equivalently, if one droplet breaks up, the sum of the volume of the two resulting droplets equals the original droplet volume. There is an implicit assumption of conservation of overall droplet volume in these coalescence and breakup processes. One may multiply Eq. (89) through by V/n, integrate over Ω_T, multiply the

resulting equation through by n and use Eq. (92) to obtain the equation of motion for the droplet volume fraction, θ_1. The result is

$$\frac{\partial \theta_1}{\partial_t} + \nabla \cdot (\theta_1 \mathbf{v}_d) = n_1 \int_{\Omega_T} \mathrm{d}V \frac{V_1}{n_1} \left[\frac{\mathrm{d}f}{\mathrm{d}t}\bigg|_c + \frac{\mathrm{d}f}{\mathrm{d}t}\bigg|_b \right] \tag{93}$$

Recalling the relationship between the local instantaneous volume fraction, ϕ_k, and the mean volume fraction, θ_k shown in Eq. (32) and in light of Eq. (27), the reason for disregarding Π_k for a dilute dispersed phase flow is evident: if the droplets are sufficiently dispersed that there are no coalesce or breakup processes to alter the number density in the droplet phase, the r.h.s. of Eq. (93) vanishes. However, Eq. (92) shows directly that the volume fraction changes not only as the number density changes, but also as the average droplet volume changes. Thus, for a reactive flow, the number density alone does not sufficiently describe the interphase conversion state because the conservation of droplet phase volume implied in this development does not hold. Droplets may change volume by virtue of evaporation, condensation or chemical reaction, in general. It is not clear, then, that Π_k should be disregarded if there is no coalescence or breakup in a given reactive flow. The task then becomes to determine under what circumstances one may disregard Π_k. It is clear that if the total volume of the dispersed phase is much much less than that of the gas phase, one may safely disregard Π_k, based *not on the no coalesce or breakup assumption, but rather on the value of the loading ratio and the size of the individual droplets*. In the FDF method for two-phase flows, it is convenient, even if not practical, to disregard $\Pi_k(\mathbf{x}, t)$ when solving the marginal TVSFMDF transport equation via Monte-Carlo methods because $\Pi_k(\mathbf{x}, t)$ makes it difficult to cast the equation in terms of a system of equivalent stochastic differential equations (SDE) (Gardiner, 1982; Pope, 1985). That is, without the non-homogeneous term, $\Pi_k(\mathbf{x}, t)$, in Eq. (27), the TVSFMDF transport equation is in the form of a (homogeneous) Fokker–Planck equation for the probability current F_L^1 (Gardiner, 1982). The principle of equivalent systems states that a given Fokker–Planck equation may yield equivalent statistics to a system of SDE's that are derived directly from that equation, thus, one may solve the system of SDE's in lieu of solving the original TVSFMDF transport equation directly (Gardiner, 1982; Pope, 1985). Generally speaking this is advantageous because the TVSFMDF transport equation is very high dimensional in each respective phase-space, and solving the TVSFMDF equation directly is difficult (Pope, 1985).

7. Conclusions

In this work, the full velocity-scalar FDF transport equation for a separated two-phase flow has been derived in the context of LES filtering for the first time. Throughout the paper, special attention has been paid to the phase-coupling terms that arise naturally in the formulation of a separated two-phase flow. Starting from the phase-filtered equations it has been shown that the instantaneous form of the transport equations for a separated two-phase flow, needed in the FDF formulation, can be recovered directly from the phase-filtered field by shrinking the LES filter volume, and are shown to be consistent with the earlier approach of Kataoka (1986). Phase-coupling terms in the context of the FDF formulation are discussed and by virtue of the formulation given herein, their functional form can be traced back to the phase-filtered formulation. The volumetric interphase conversion term, which balances the total time rate of change of the local instantaneous volume fraction, has been analyzed and conditions under which it may be disregarded discussed. Further, it is shown that conditionally surface-filtered quantities arising in the two-phase FDF formulation are equal (within a factor of density filtered on the surface) to phase-coupling terms in the phase-filtered LES equations. The benefit of this result is that existing closures for phase-filtered coupling terms, which are in general problem specific, may be directly substituted into the TVSFMDF transport equation. This will be the focus of future research.

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