QUADRATURE-BASED MOMENT METHODS

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INTRODUCTION

Many of the models used in chemical engineering involve distribution functions. Examples include particle size distributions in multiphase flows, molecular weight distributions in polymer systems, and probability distributions in turbulent flows. In general, transport equations for such distribution functions arise naturally in mesoscale descriptions of microscale phenomena, the classical example being the Boltzmann equation of molecular transport and collisions in an ideal gas. Another classical example is the Smolokowski equation for Brownian aggregation of nanoparticles that describes the number density of particles composed of a given number of monomers. In other applications such as aerosol transport, the distribution function will have more than one internal variable (e.g., volume and surface area). Indeed, the number of internal variables can be quite large when many microscale properties evolve at the same time. The resulting transport equation for the distribution function will have a large number of independent variables (time, physical space, internal variables) and its solution for realistic flow geometries (e.g., a chemical reactor) represents a significant challenge.

In the chemical engineering literature, the transport equation for the distribution function is usually referred to as a population balance equation (PBE). However, it is important to distinguish between a *dynamically passive* distribution function wherein the velocity does not appear as an internal variable, and a *dynamically active* distribution function (e.g., the velocity distribution in the Boltzmann equation). For the latter, the internal variable (i.e., velocity) determines the spatial transport, while in the former the spatial transport occurs due to an external velocity field (e.g., the fluid velocity) that is found by solving a separate momentum equation. In order to make this distinction clear, we use the term "PBE" to refer to the transport equation for a dynamically passive distribution function, and the term "generalized PBE" (GPBE) to refer to the transport equation for a dynamically active distribution function. Note that many of the applications of distribution functions in chemical engineering (e.g., colloids, aerosols, nanoparticles) involve very small particles whose mean velocity is the same as the carrier fluid, and thus they can be described by a PBE. However, for larger particles with non-zero Stokes numbers (e.g., granular flows, gas-solid flows, bubbly flows) it is necessary to use the GPBE. In the more complicated case of polydisperse multiphase flows, both the particle size and the particle velocity appear in the GPBE as internal variables. Likewise, for polydisperse reacting multiphase flows additional internal variables are needed to describe the chemical composition and temperature.

The numerical solution of a PBE or a GPBE is challenging due to the number of independent variables. Only in the simplest cases, where the distribution function is spatially homogeneous and contains one or, at most two, internal variables, is a direct solution possible. Otherwise, for modeling spatially inhomogeneous cases (e.g. a gas-solid riser), it is necessary to reduce the number of degrees of freedom used to represent the distribution function. The classical Eulerian method for accomplishing this task is to solve transport equations for a finite set of moments of the distribution function [1]. For example, in the kinetic theory of an ideal gas near equilibrium, the Boltzmann equation for the velocity distribution function is replaced by transport equations for density (zero-order moment), mean velocity (first-order moments), and energy (trace of second-order moments). However, for rarefied gases, closure at the level of the second-order moments is no longer sufficient [2]. More generally, the transport equations for the moments derived from the PBE or GPBE will not be closed for a finite set. It is thus

necessary to introduce a *closure* that prescribes how the moments not included in the set of transported moments are found from the transported moments. This procedure is not uniquely defined and thus, ideally, we would like to find a closure that minimizes the error in the unclosed moments. The quadrature method of moments is such a procedure that, for univariate distribution functions on finite intervals, has minimum error and tight error bounds [3, 4]. For univariate distribution functions, an efficient algorithm exists [3, 4, 5] for computing the quadrature weights and abscissas given a finite set of moments. Solution methods based on inverting a finite set of moments to compute weights and abscissas are referred to as *quadrature-based moment methods*. For strongly non-equilibrium flows (e.g., wherein collisions are rare), the most reliable method for approximating the moments of a distribution function is to estimate them from a Lagrangian simulation. However, such estimates have statistical noise and can be inaccurate for two-way coupling between the solid and fluid phases. In comparison, Eulerian methods are straightforward for two-way coupling and generally less costly for the same level of accuracy.

APPLICATIONS OF QUADRATURE-BASED MOMENT METHODS

The quadrature method of moments (QMOM) were first applied by McGraw [6] to close a univariate PBE equation for an aerosol undergoing coagulation. A multi-variate PBE can be treated in a similar manner using the direct quadrature method of moments (DQMOM) described in [7, 14]. The treatment of more complicated particle processes described by a PBE for nucleation and growth coupled with aggregation is relatively straightforward and surprisingly accurate given the low computational cost [8, 9, 10]. Quadrature-based moment methods are particularly attractive for solving a PBE coupled to a computational fluid dynamics (CFD) code [10, 17]. A similar approach can also be used to treat conditional moments with good accuracy [16]. Because of the velocity variable, the treatment of a GPBE requires special care. The simplest applications can be treated as *mono-kinetic* (i.e., all particle with the same size have the same velocity), resulting in an Eulerian multi-fluid model [20] where only the mean velocity conditioned on the particle size appears [11, 12, 13]. In essence, such problems are treated by using quadrature-based moment methods for the size moments without accounting for correlations with the velocity. The range of validity of the mono-kinetic assumption is however rather limited, holding primarily for particles with low Stokes numbers and when particles do not cross each other. To do better, one must use quadrature-based moment methods to treat GPBE moments involving velocity [15, 18, 19] or joint moments involving size and velocity [20]. It is then possible to compute jets of non-colliding particles and particles with large Stokes numbers - something that cannot be done with standard two-fluid models.

The treatment of velocity moments requires an efficient moment-inversion algorithm to find the weights and three components of the velocity abscissas for each quadrature node starting from the moments. Mathematically, this is a non-trivial task and current algorithms [21, 23] use multiple univariate quadratures [4] to construct tensor-product abscissas and non-negative weights. An alternative would be to use DQMOM with optimal moment sets [22]; however, care must be taken to handle cases where the velocity abscissas have shocks and other non-smooth behavior. Remarkably, treating shocks and other highly a non-equilibrium behavior with QMOM is straightforward [21, 23]. The fully coupled treatment of fluid-solid flows uses a GPBE for the solid phase and a hydrodynamic equation for the fluid phase [24]. The coupling between the phases occurs at the level of the velocity moments. Thus, unlike with Euler-Lagrange description, quadrature-based moment closures for multiphase flows are fully Eulerian and do not suffer from statistical noise due to finite sample sizes. Ultimately, it should be possible to develop an Eulerian CFD code for polydisperse multiphase flows

that can capture highly non-equilibrium behavior (e.g., jet crossing, wall rebounds, Knudsen layers) currently captured only by Lagrangian solvers.

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