Modeling of stirred liquid-liquid dispersions *

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March 4, 2005

Abstract

In this paper, we consider a stirred liquid-liquid dispersion, i. e. a tank filled with two immiscible fluids which are stirred so that one of the phases disperses into the other one by building droplets.

To model all relevant processes appearing in such a system, one has to account for the turbulent flow in the tank as well as for the population dynamical processes of the dispersed phase. We derive a system of equations that contains both occurring phenomena.

Furthermore, the properties of the corresponding differential-algebraic equations describing the dynamics of the process will be determined in order to analyze the behavior of the system when solving it numerically.

1 Introduction

In this report, we consider a stirred tank filled with two immiscible fluids (such as water and oil, for example). When the two fluids are stirred, one disperses into the other one by building droplets (see Fig. 1). All droplets together are called the dispersed phase, whereas the other fluid is called the continuous phase.

To model the phenomena that occur in the stirred tank, one has to describe the turbulent flow of the continuous phase as well as the behavior of the dispersed phase. Therefore, one has to account for the population dynamical processes of the drops, i. e. one has to describe how the number of drops of a certain size changes with time. On the one hand, when two drops collide, they may form a larger drop. That means a new larger drop is generated, whereas two smaller drops disappear. On the other hand, a drop may also collide with an eddy such that it breaks up into some smaller drops. In this case, some smaller drops are formed and the larger, so-called mother drop, is destroyed. The confluence of

*Thanks to the DFG-Graduiertenkolleg "Transport Phenomena with moving boundaries" http://fspfst.fluidsystemtechnik.tu-berlin.de/fst/KollegSite/en/index.html

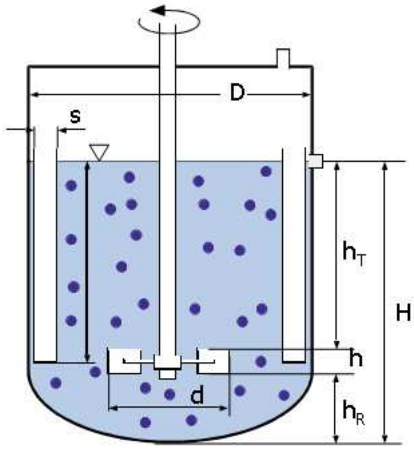

Figure 1: Stirred liquid-liquid system

drops is called coalescence, the break-up is called dispersion.
Note that we only consider binary coalescence (since the probability that three or more drops collide in a time interval $(t, t+dt)$ at a certain point is negligibly small compared to the probability that two drops meet), but allow dispersion of a drop into two or more daughter drops. The two processes can be seen in Fig. 2.

In the next section, we will derive a mathematical model describing the above mentioned processes. The flow field in the stirred tank can be described by the Navier-Stokes equations for incompressible fluids. Furthermore, we will model turbulence by Reynolds-averaging and then solve the arising closure problem by using a k-ε model. The behavior of the drops is modeled by a population balance equation, where coalescence and dispersion appear as source and sink terms on the right-hand side.

Then, in Section 3, the system of equations will be discretized in space in order to determine the index of the underlying differential-algebraic system that describes the dynamics of the process. Generally, one can say that the higher the index, the more difficult it is to solve the problem numerically [BCP89]. Therefore, it is quite important to know the index so that the problem can be treated appropriately. If the index is higher, index reduction may be one way to treat the problem.

In Appendix A, we briefly discuss the phenomena of coalescence and dispersion. There, we also derive some physically based formulas to model these processes mathematically.

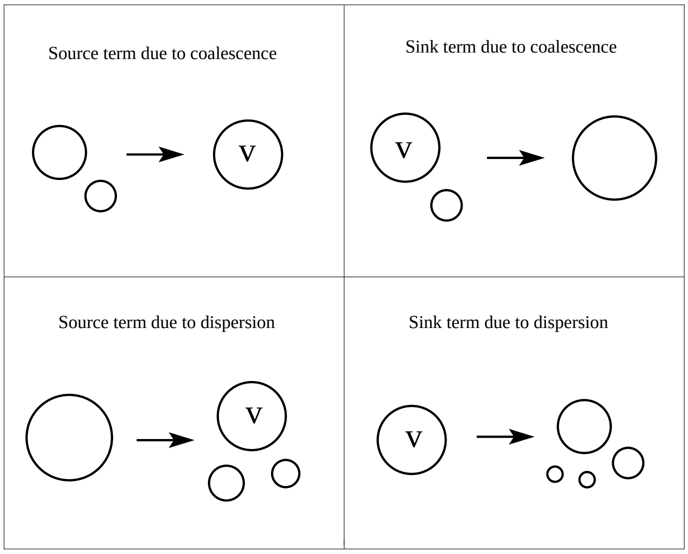

Figure 2: Source and sink terms for a drop of volume V due to coalescence and dispersion

In Appendix B, we give a short introduction to the concept of the strangeness-index for nonlinear differential-algebraic equations.

2 Modeling

If we want to model the processes that occur in a stirred tank appropriately, we do not only need spatial coordinates $x = (x_1, x_2, x_3)^T \in \Omega \subset \mathbb{R}^3$ and a time coordinate $t \in [0, T]$ but also so-called internal coordinates $e = (e_1, \ldots, e_m)^T \in \mathbb{R}^m$, which describe m different properties of the dispersed phase. We need these additional coordinates, since the individuals of the dispersed phase (i. e. the drops) must be distinguishable from each other with respect to some significant characteristics.

In general, these internal coordinates describe properties related to geometry (e. g., size, volume), material properties (e. g., density, chemical composition, color), or characteristics in the interaction with the continuous phase (e. g., surface charge).

In the discussed application, it is sufficient to distinguish the drops due to their size. Therefore, in this paper, we only use one internal coordinate, namely the volume V. Note that, in the literature, there is often used the diameter d of a drop as internal coordinate. Therfore, most of the formulas in Appendix A are also given for the diameter. But since the diameter d of a drop is directly related to its volume V by the relation $V = c_V d^3$, the formulas from the appendix can easily be transformed such that they depend on the volume V. (For spherical drops one has: $c_V = \frac{\pi}{6}$; for other shapes c_V has to be chosen appropriately.)

In the simulation package Parsival [Wul], which is used for the solution of the population balance equation within our project, the volume as well as the diameter can be chosen as internal coordinate. However, some modules are harder to implement if the volume is used. The coordinate transformations, which describe how to switch between volume and diameter representation in the different terms of the population balance equation, can be found in the Parsival tutorial [GB04].

In this context, when internal coordinates are used, the space coordinates are called external coordinates accordingly.

2.1 Modeling the flow

First, we model the flow in the stirred tank. We assume that the drops do not have any influence on the flow field. (They are just moving with the fluid with the same velocity.) This implies that the description of the flow is independent of the internal coordinates.

The flow field in the stirred tank is described by the Navier-Stokes equations for incompressible fluids [GDN95]

$$\left.\begin{array}{rcl} \frac{\partial}{\partial t}u + \nabla \cdot (u \otimes u) + \frac{1}{\rho_f}\nabla p & = & \nu \Delta u + g \\ \nabla \cdot u & = & 0 \end{array}\right\} \quad \text{in } \Omega, \tag{1}$$

where u is the velocity of the fluid, p is the pressure, and g are the external forces. In the discussed application, the only external force that has to be considered is gravity. Furthermore, ρ_f is the density of the fluid, $\nu = \frac{\mu}{\rho_f}$ is the kinematic viscosity, and μ is the dynamic viscosity. The first equation in (1) accounts for the conservation of momentum. Here, $\frac{\partial}{\partial t}u + \nabla \cdot (u \otimes u)$ describes the acceleration of the particles of the fluid, ∇p is the pressure gradient, and $\nu \Delta u$ accounts for the friction between the particles of the fluid. The second equation in (1) is called the "continuity equation" and models the conservation of mass.

Additionally, we prescribe the following boundary conditions on $\partial\Omega =: \Gamma = \Gamma_{\text{stirrer}} \cup \Gamma_{\text{wall}} \cup \Gamma_{\text{surface}}$:

$$u = u_{\text{stirrer}} \ \text{ on } \Gamma_{\text{stirrer}}, \quad u = 0 \ \text{ on } \Gamma_{\text{wall}}, \quad \frac{\partial u_1}{\partial \mathfrak{n}_1} = \frac{\partial u_2}{\partial \mathfrak{n}_2} = 0, \ u_3 = 0 \ \text{ on } \Gamma_{\text{surface}},$$

where Γ_{stirrer} describes the stirrer, Γ_{wall} the fixed walls, and Γ_{surface} the surface of the liquid in the tank. Furthermore, u_1, u_2, and u_3 are the velocities in x_1-,

x_2-, and x_3-direction, and $\mathfrak{n}_1$ and $\mathfrak{n}_2$ are the outer normal vectors in x_1- and x_2-direction, respectively. The prescribed velocity on the stirrer u_{stirrer} is given by

$$u_{\text{stirrer}} = \left[-r\sin(\varphi)\omega, r\cos(\varphi)\omega, 0\right]^T,$$

where the radius r and the angle φ are defined by

$$r = \sqrt{x_1^2 + x_2^2}, \quad \varphi = \arccos\left(\frac{x_1}{r}\right),$$

and ω is the constant frequency. This frequency is determined by the adjusted rotational speed N^*. This parameter can be used as a control input in order to influence the drop size distribution.
The initial condition is chosen such that it is consistent with the boundary conditions.

The turbulence of the flow is modeled by "averaging" each quantity. The idea is that in most practical investigations of turbulent flows, one is not interested in every microscopic detail but only in macroscopically observed mean values [GDN95]. That is why we split each quantity – velocity u, pressure p, and outer forces g – into a mean part $\overline{u}$, $\overline{p}$, and $\overline{g}$, respectively, and into the so-called fluctuations u', p', and g', which model smallest variations of each quantity:

$$u = \overline{u} + u', \quad p = \overline{p} + p', \quad g = \overline{g} + g'.$$

The mean part is usually chosen as a component-wise temporal averaged value. But in general also other averages can be used. This can be expressed by applying a filter $\langle\cdot\rangle$, i. e.

$$\overline{u} := \langle u\rangle, \quad \overline{p} := \langle p\rangle, \quad \overline{g} := \langle g\rangle.$$

After introducing these relations into the Navier-Stokes equations (1) and averaging them (i. e. applying the filter $\langle\cdot\rangle$ to them) we get the Reynolds-averaged Navier-Stokes equations

$$\left.\begin{array}{rcl} \frac{\partial}{\partial t}\overline{u} + \nabla\cdot(\overline{u}\otimes\overline{u}) + \frac{1}{\rho_f}\nabla\overline{p} - \nu\Delta\overline{u} + \nabla\cdot\langle u'\otimes u'\rangle & = & \overline{g} \\ \nabla\cdot\overline{u} & = & 0 \end{array}\right\} \quad \text{in } \Omega,$$

where the tensor

$$R(u') := -\langle u'\otimes u'\rangle$$

is known as Reynolds stress tensor. This shows that the averaging leads to almost the same equations as we had before, just with the additional term $-\nabla\cdot R(u')$. Since R depends on the fluctuations u' of the velocities, the system consists of one more unknown variable u' such that the Reynolds averaged Navier-Stokes equations do not build a closed system any more, i. e. there are more unknowns than equations. This difficulty is known as the closure problem in turbulence modeling [GDN95].

In order to solve this problem, one has to introduce new equations which are usually based on hypotheses and approximations given by empirical information or experimental data. These equations form the so-called "turbulence model".

The most widely used model is the k-ε model by Launder and Spalding [LS72]. It is a so-called two-equations model, since two additional partial differential equations are introduced in order to get a closed system. In the k-ε model, the following two additional variables are used to model the Reynolds stresses: the turbulent kinetic energy k and its dissipation rate ε given by

$$k := \frac{1}{2}\langle \|u'\|_F \rangle, \quad \varepsilon := \frac{\nu}{2}\langle \|\nabla u' + (\nabla u')^T\|_F^2 \rangle,$$

where $\|\cdot\|_F$ denotes the Frobenius norm defined by

$$\|X\|_F^2 = \mathrm{tr}(XX^T) = \sum_{i=1}^{n}\sum_{j=1}^{m} x_{ij}^2$$

for matrices $X = [x_{ij}]_{i=1,\dots,n,j=1,\dots,m} \in \mathbb{R}^{n\times m}$. Note that this definition also includes the norm of a vector $x \in \mathbb{R}^n$, since it can be seen as a $n \times 1$-matrix.

In the k-ε model the Reynolds tensor $R(u')$ is approximated by

$$R(u') \approx \mathcal{R}(\nabla\overline{u}, k, \varepsilon) := -\frac{2}{3}kI + c_\mu \frac{k^2}{\varepsilon}(\nabla\overline{u} + (\nabla\overline{u})^T),$$

where I denotes the identity matrix. If we insert this into the averaged momentum equation and set $\nu^* := \nu + \nu_t$ with

$$\nu_t = c_\mu \frac{k^2}{\varepsilon}, \tag{2}$$

we get

$$\frac{\partial}{\partial t}\overline{u} + \nabla\cdot(\overline{u}\otimes\overline{u}) + \frac{1}{\rho_f}\nabla\overline{p} + \frac{2}{3}\nabla k - \nabla\cdot(\nu^*(\nabla\overline{u} + (\nabla\overline{u})^T)) = \overline{g}.$$

With the two additional transport equations for k and ε, which close the system of equations, the following system is derived:

$$\begin{aligned}
\frac{\partial}{\partial t}\overline{u} + \nabla\cdot(\overline{u}\otimes\overline{u}) + \frac{1}{\rho_f}\nabla\overline{p} + \frac{2}{3}\nabla k - \nabla\cdot(\nu^*(\nabla\overline{u} + (\nabla\overline{u})^T)) &= \overline{g}, \\
\frac{\partial}{\partial t}k + \overline{u}\cdot\nabla k - \frac{\nu_t}{2}\|\nabla\overline{u} + (\nabla\overline{u})^T\|_F^2 - \nabla\cdot(\nu_t\nabla k) + \varepsilon &= 0, \\
\frac{\partial}{\partial t}\varepsilon + \overline{u}\cdot\nabla\varepsilon - \frac{c_1 c_\mu}{2}k\|\nabla\overline{u} + (\nabla\overline{u})^T\|_F^2 - \nabla\cdot\left(\frac{1}{c_\varepsilon}\nu_t\nabla\varepsilon\right) + c_2\frac{\varepsilon^2}{k} &= 0,
\end{aligned}$$

where the constants $c_\mu, c_\varepsilon, c_1$ and c_2 are chosen from experimental investigations. Launder and Spalding [LS72] proposed the following values:

$$c_\mu = 0.09, \quad c_\varepsilon = 1.3, \quad c_1 = 1.44, \quad c_2 = 1.92.$$

For the averaged velocity $\overline{u}$ we can use the same boundary conditions as prescribed for the velocity u in the Navier-Stokes equations (1) before. Therefore, we set:

$$\overline{u} = u_{\text{stirrer}} \text{ on } \Gamma_{\text{stirrer}}, \quad \overline{u} = 0 \text{ on } \Gamma_{\text{wall}}, \quad \frac{\partial \overline{u}_1}{\partial \mathfrak{n}_1} = \frac{\partial \overline{u}_2}{\partial \mathfrak{n}_2} = 0, \ \overline{u}_3 = 0 \text{ on } \Gamma_{\text{surface}}$$

with the same notation as above.

For k and ε we additionally prescribe the following boundary conditions:

$$k = 0 \text{ on } \Gamma_{\text{stirrer}} \cup \Gamma_{\text{wall}}, \quad \frac{\partial k}{\partial \mathfrak{n}} = 0 \text{ on } \Gamma_{\text{surface}},$$

and

$$\frac{\partial \varepsilon}{\partial \mathfrak{n}} = 0 \text{ on } \Gamma,$$

where $\mathfrak{n}$ denotes the outer normal vector. Again, the initial conditions for $\overline{u}$, k, and ε are chosen such that they are consistent with the boundary conditions.

2.2 Modeling the dispersed phase

The population dynamical processes can be modeled using the following general population balance equation given in Gerstlauer [Ger99]:

$$\frac{\partial f(e,x,t)}{\partial t} = -\nabla_{x,e} \cdot \phi_N(e,x,t) + s(e,x,t),$$

where $f(e,x,t)$ is the number density function depending on internal and external coordinates e and x, respectively, as well as on time t. Further, $\nabla_{x,e}\cdot$ denotes the divergence operator with respect to internal and external coordinates. The right-hand side consists of the transport density $\nabla_{x,e} \cdot \phi_N$ and the sum of source and sink terms s.

The transport density $\nabla_{x,e} \cdot \phi_N(e,x,t)$ can be split up into the transport density concerning the internal coordinates $\nabla_e \cdot \phi_{N,e}$ and the one concerning the external coordinates $\nabla_x \cdot \phi_{N,x}$:

$$\nabla_{x,e} \cdot \phi_N(e,x,t) = \nabla_x \cdot \phi_{N,x}(e,x,t) + \nabla_e \cdot \phi_{N,e}(e,x,t),$$

where the ∇-operators are defined by

$$\nabla_x := \begin{bmatrix} \frac{\partial}{\partial x_1} \\ \frac{\partial}{\partial x_2} \\ \frac{\partial}{\partial x_3} \end{bmatrix} \quad \text{and} \quad \nabla_e := \begin{bmatrix} \frac{\partial}{\partial e_1} \\ \vdots \\ \frac{\partial}{\partial e_m} \end{bmatrix}.$$

Furthermore, the transport flows $\phi_{N,x}$ and $\phi_{N,e}$ can be divided into a convective and a diffusive part:

$$\begin{aligned} \phi_{N,x} &= w_x \cdot f(e,x,t) + \phi_{N,x}^D, \\ \phi_{N,e} &= w_e \cdot f(e,x,t) + \phi_{N,e}^D. \end{aligned}$$

The convective part in x-direction $w_x \cdot f(e,x,t)$ describes the deterministic movement of the particles. The diffusive part $\phi^D_{N,x}$ can be used to account for the Brownian motion of the particles. The convective part in direction of the internal coordinates states the rate of change of the single particle properties. The diffusive part can be used to describe the stochastic variation of the rate of change.

In the discussed application, we consider a system without changes in the internal coordinates per time, i. e. there is no growth or shrinkage of droplets due to mass transfer or reaction. Therefore, we get $\nabla_e \cdot \phi_{N,e}(e,x,t) = 0$. Furthermore, the Brownian motion is assumed to be very small compared to the deterministic particle movement and therefore will be neglected. Additionally, we can approximate the velocity w_x by the velocity u of the surrounding continuous phase. (The drops are just moving with the surrounding fluid.)

With these simplifications and using just one internal coordinate, namely the volume V, we get the following population balance equation:

$$\frac{\partial f(V,x,t)}{\partial t} = -\nabla_x \cdot (u \cdot f(V,x,t)) + s(V,x,t). \tag{3}$$

Note that the divergence operator $\nabla_x \cdot$ used here is equal to the divergence operator $\nabla \cdot$ in the Navier-Stokes equations (1), since we do not have any derivatives with respect to the internal coordinate V in both cases. Therefore, we will in the following just use $\nabla \cdot$ instead of $\nabla_x \cdot$.

For f we prescribe the following initial condition

$$f(V,x,t=0) = f_0(V,x),$$

where – for simplicity – f_0 is for fixed x and t usually chosen as the density of a Gaussian normal distribution. Additionally, we use as boundary condition

$$f(V,x,t) = 0 \text{ on } \Gamma.$$

Furthermore, s is the sum of source and sink terms describing the population dynamical processes. In our application only coalescence and dispersion can be observed. Therefore, the term s can be divided into

$$s = s^+_{\text{coal}} + s^-_{\text{coal}} + s^+_{\text{disp}} + s^-_{\text{disp}},$$

where $s^{\pm}_{\text{coal}}$ and $s^{\pm}_{\text{disp}}$ are describing the rate of increase / decrease of the number density function f due to coalescence and dispersion, respectively. The four different terms occurring on the right-hand side are explained graphically in Fig. 2. In the following, we discuss the mathematical representation of these terms.

Let us start with the coalescence process:
The source term s^+_{coal} due to coalescence accounts for all the drops which are

formed by coalescence of two smaller drops. So, the source term can be written as

$$s^{+}_{\text{coal}}(V,x,t) = \int_0^V R_{\text{coal}}(V',V'',y(x,t))f(V',x,t)f(V'',x,t)dV',$$

where V' and V'' are the volumes of the two coalescing drops. Since the drops with volumes V' and V'' form a drop with volume V, the following relation must hold:

$$V'' = V - V'.$$

The function R_{coal} describes the probability that two individuals coalesce. The vector y denotes the so-called continuous phase vector (e. g. [Ram00]), i. e. the properties of the continuous phase that influence the coalescence process. Thus, the vector y may consist of pressure, temperature, or other values that we get from the calculation of the flow field.
Note that the lower limit of the integral could also be substituted by the minimal possible volume of a drop $V_{\min}$.

The sink term due to coalescence accounts for the droplets that are lost because they form a larger one together with another droplet. Mathematically, the sink term due to coalescence is given by integration over all drops which are able to coalesce with a given one:

$$s^{-}_{\text{coal}}(V,x,t) = -f(V,x,t)\int_0^{V_{\max}-V} R_{\text{coal}}(V,V',y(x,t))f(V',x,t)dV'.$$

Dispersion can, like coalescence, mathematically be modeled by a source and a sink term. The source term describes the generation of daughter droplets while the sink term stands for the loss of the mother drop.

The source term due to dispersion is given by

$$s^{+}_{\text{disp}}(V,x,t) = \int_V^{V_{\max}} n(V',y(x,t))\gamma(V,V',y(x,t))R_{\text{disp}}(V',y(x,t))f(V',x,t)dV',$$

where $n(V',y(x,t))$ is the number of daughter drops formed by dispersion of a drop with volume V', $\gamma(V,V',y(x,t))$ is the probability density function that describes the probability that a daughter drop with volume V is formed by the dispersion of a mother drop with volume V', and $R_{\text{disp}}(V',y(x,t))$ is the breakage rate that accounts for the number of dispersed drops per unit time. The function γ has to fulfill the following normalization condition:

$$\int_0^{V_{\max}} \gamma(V,V',y(x,t))dV = 1. \tag{4}$$

Conservation of mass leads to the additional condition:

$$n(V',y(x,t))\int_0^{V'} m(V)\gamma(V,V',y(x,t))dV = m(V'), \tag{5}$$

where $m(V)$ denotes the mass of a drop with volume V.

The sink term due to dispersion accounts for the drops which are lost because they break up into smaller droplets. It is given by

$$s_{\text{disp}}^{-}(V,x,t) = -R_{\text{disp}}(V,y(x,t))\, f(V,x,t).$$

The coalescence rate R_{coal}, the dispersion rate R_{disp}, the distribution of daughter drops γ as well as the number of daughter drops n have to be specified for certain dispersion processes depending on the considered application.

In Appendix A, we present some different approaches how these functions can be modeled in our application. There, we consider three different approaches: one by Coulaloglou and Tavlarides [CT77], one by Tsouris and Tavlarides [TT94], and one by Ritter [Rit02]. In all these models, it is assumed that the dispersed phase is homogeneously distributed in the physical space, i. e. that we have an ideally mixed tank. In this case, a space-independent population balance equation can be used to describe the processes in the stirred tank. In addition, there does not occur any transport term in their population balance equations but only changes of the number density function due to coalescence and dispersion. But although we use a more general approach (namely a space-dependent number density function $f(V,x,t)$ and changes in f due to convective transport, see Eq. (3)), we can nevertheless use the coalescence and dispersion rates proposed there. In our model, we account for the space-dependency in the coalescence and dispersion rates by substituting the constant value ε_l used in the three mentioned papers by the local energy dissipation rate ε (from the k-ε model) at a certain point (x,t).

To model turbulence, the population balance equation is also averaged. We use the same approach as for the Navier-Stokes equations, i. e. we split all occurring quantities, which here are the number density function f and the velocity u, into mean values $\overline{f}$ and $\overline{u}$, and fluctuations f' and u'. Again, we regard $\overline{f}$ and $\overline{u}$ as averaged values, received by a filter operator $\langle\cdot\rangle$

$$\begin{aligned} f &= \overline{f} + f', \quad \overline{f} = \langle f \rangle, \\ u &= \overline{u} + u', \quad \overline{u} = \langle u \rangle. \end{aligned}$$

Inserting this into Eq. (3), we get

$$\frac{\partial(\overline{f}+f')}{\partial t} = -\nabla\cdot((\overline{u}+u')(\overline{f}+f')) + s.$$

Applying the filter operator $\langle\cdot\rangle$ to this equation, we then get the averaged population balance equation

$$\frac{\partial\overline{f}}{\partial t} = -\nabla\cdot(\overline{u}\overline{f}) - \nabla\cdot(u'f') + \overline{s},$$

where $\overline{s}$ is the mean value of s, i. e. $\overline{s} = \langle s \rangle$. This term represents the averaged source and sink terms.

The term $\nabla \cdot (u'f')$ is modeled by

$$\nabla \cdot (u'f') = -\nabla \cdot (c_t \nabla \overline{f})$$

in analogy to the approach for the Reynolds stress tensor (see [Pas04]). Similar to the Reynolds stress tensor, which is assumed to be proportional to the gradient of the velocity in this approach, it is assumed here that the term $(u'f')$ is proportional to the gradient of the averaged number density function $\nabla \overline{f}$. Furthermore, c_t is specified by the turbulent Schmidt number Sc_t

$$\mathrm{Sc}_t = \frac{\mu_t}{\rho_f c_t},$$

which is assumed to be approximately one. (In many applications the value 0.9 is used.)
The turbulent eddy viscosity μ_t is given by the turbulent kinetic energy k and the energy dissipation rate ε (cf. Eq. (2)):

$$\mu_t = \nu_t \rho_f = c_\mu \rho_f \frac{k^2}{\varepsilon}.$$

Together, the averaged population balance equation then reads as

$$\frac{\partial \overline{f}}{\partial t} = -\nabla \cdot (\overline{u}\overline{f}) + \nabla \cdot (c_t \nabla \overline{f}) + \overline{s}. \tag{6}$$

Again, we can apply the former initial and boundary conditions for f to our averaged number density function $\overline{f}$.

3 Analysis of the differential-algebraic system

In order to analyze the dynamics of the system, we apply a method of lines (MOL) approach to the system of equations describing the considered stirred tank reactor. This spatial discretization leads to a differential-algebraic system, for which the index will be determined.

3.1 Derivation of the differential-algebraic system

As we have seen in the last section, the system of equations describing the processes in the stirred tank is given by:

$$\left.\begin{array}{rcl} \frac{\partial}{\partial t}\overline{u} + \nabla \cdot (\overline{u} \otimes \overline{u}) + \frac{1}{\rho_f}\nabla \overline{p} + \frac{2}{3}\nabla k - \nabla \cdot (\nu^* (\nabla \overline{u} + (\nabla \overline{u})^T)) & = & \overline{g}, \\ \frac{\partial}{\partial t}k + \overline{u} \cdot \nabla k - \frac{\nu_t}{2}\|\nabla \overline{u} + (\nabla \overline{u})^T\|_F^2 - \nabla \cdot (\nu_t \nabla k) + \varepsilon & = & 0, \\ \frac{\partial}{\partial t}\varepsilon + \overline{u} \cdot \nabla \varepsilon - \frac{c_1}{2}k\|\nabla \overline{u} + (\nabla \overline{u})^T\|_F^2 - \nabla \cdot \left(\frac{c_\varepsilon}{c_\mu}\nu_t \nabla \varepsilon\right) + c_2 \frac{\varepsilon^2}{k} & = & 0, \\ \frac{\partial \overline{f}}{\partial t} + \nabla \cdot (\overline{u}\overline{f}) - \nabla \cdot (c_t \nabla \overline{f}) & = & \overline{s}, \\ \nabla \cdot \overline{u} & = & 0 \end{array}\right\} \tag{7}$$

with constants $c_\mu = 0.09$, $c_\varepsilon = 0.07$, $c_1 = 0.126$, and $c_2 = 1.92$ as defined in Section 2.
Note that $\nu^* = \nu + \nu_t$ with $\nu_t = c_\mu \frac{k^2}{\varepsilon}$ and $c_t = \frac{\mu_t}{\mathrm{Sc}_t} = \frac{c_\mu \rho_f \frac{k^2}{\varepsilon}}{\mathrm{Sc}_t}$ are dependent on the quantities k and ε. Furthermore, since the term s is modeled in such a way that it is dependent on the local energy dissipation rate ε (see Appendix A), the averaged source term $\overline{s}$ is also dependent on ε.
Additionally, we have to take care that the condition for mass conservation, given by Eq. (5), is fulfilled.

In the following, the system of equations given in (7) will be discretized in space in order to analyze the resulting differential-algebraic system.
In analogy to the construction in [BCP89], it can be shown that the spatial discretization of the Navier-Stokes equations (1) leads to a semi-discretized system of the following form:

$$\begin{aligned} M\frac{du_h}{dt} + (K + N(u_h))u_h + \frac{1}{\rho_f}Cp_h &= g_h, \\ \tilde{C}^T u_h &= 0, \end{aligned}$$

where u_h, p_h, and g_h are the semi-discretized velocity, pressure, and outer forces, respectively. Further, M is the mass matrix and K and N are the discretized diffusive and convective parts, corresponding to $-\nu\Delta u$ and $\nabla\cdot(u\otimes u)$, respectively. The matrices C and $\tilde{C}^T$ are the discretized gradient and divergence operators. (Normally the discretization is done in such a way that $\tilde{C} = C$ holds, but in general this need not be like this.)

For the system in (7), consisting of the Reynolds-averaged Navier-Stokes equations with k-ε model and the averaged population balance equation, this can be done in the same way. Here, spatial discretization leads to

$$\left.\begin{aligned} M_1\tfrac{du_h}{dt} + (N_1(u_h) + N_2(k_h,\varepsilon_h))u_h + \tfrac{2}{3}C_1k_h + \tfrac{1}{\rho_f}C_2p_h &= \mathcal{G}_1, \\ M_2\tfrac{dk_h}{dt} + (N_3(u_h) + N_4(k_h,\varepsilon_h))k_h + N_5(k_h,\varepsilon_h)u_h &= \mathcal{G}_2(\varepsilon_h), \\ M_3\tfrac{d\varepsilon_h}{dt} + (N_3(u_h) + \tilde{N}_4(k_h,\varepsilon_h))\varepsilon_h + \tilde{N}_5(k_h,\varepsilon_h)u_h &= \mathcal{G}_3(k_h,\varepsilon_h), \\ M_4\tfrac{df_h}{dt} + (N_6(u_h) + N_7(k_h,\varepsilon_h))f_h &= \mathcal{G}_4(\varepsilon_h,f_h), \\ C_3^T u_h &= 0, \end{aligned}\right\} \tag{8}$$

where u_h and p_h are the semi-discretized averaged velocity and pressure, respectively. Furthermore, k_h, ε_h, and f_h are the corresponding quantities derived by the semi-discretization of the turbulent kinetic energy k, the energy dissipation ε, and the averaged number density function $\overline{f}$. The terms $\mathcal{G}_1, \ldots, \mathcal{G}_4$ on the right-hand side are the semi-discretized source terms. The matrices $M_1, \ldots, M_4$ are mass matrices, the matrices $N_1, \ldots, N_7$, $\tilde{N}_4$ and $\tilde{N}_5$ are discretizations of the nonlinear terms. Finally, C_1 and C_2 are the discretized gradient operators and C_3^T is the discretized divergence operator.

Additionally, the discretization of the equation for mass conservation, given by Eq. (5), leads to

$$\mathcal{G}_5(m_h) = 0, \tag{9}$$

where m_h is the vector consisting of the discretized particle masses.

To make the notation clearer, we summarize the quantities u_h, k_h, ε_h, and f_h to the variable $q_h := [u_h^T, k_h^T, \varepsilon_h^T, f_h^T]^T$. With this, the system in (8) together with Eq. (9) can be written as

$$\begin{bmatrix} M & & \\ & 0 & \\ & & 0 \end{bmatrix} \begin{bmatrix} \dot{q}_h \\ \dot{m}_h \\ \dot{p}_h \end{bmatrix} = \begin{bmatrix} \mathcal{G} + \mathcal{P} \\ \mathcal{G}_5 \\ C_3^T u_h \end{bmatrix}, \tag{10}$$

where $\dot{q}_h$, $\dot{m}_h$, and $\dot{p}_h$ denote the time derivatives of the corresponding variables. Furthermore, M, $\mathcal{G}$ and $\mathcal{P}$ are given by

$$M = \begin{bmatrix} M_1 & & \\ & \ddots & \\ & & M_4 \end{bmatrix}, \quad \mathcal{G} = \begin{bmatrix} -(N_1 + N_2)u_h - \frac{2}{3}C_1 k_h + \mathcal{G}_1 \\ -(N_3 + N_4)k_h - N_5 u_h + \mathcal{G}_2 \\ -(N_3 + \tilde{N}_4)\varepsilon_h - \tilde{N}_5 u_h + \mathcal{G}_3 \\ -(N_6 + N_7)f_h + \mathcal{G}_4 \end{bmatrix},$$

and

$$\mathcal{P} = \begin{bmatrix} -\frac{1}{\rho_f} C_2 p_h \\ 0 \\ 0 \\ 0 \end{bmatrix}.$$

Note that $\mathcal{G}$ is not dependent on the semi-discretized pressure p_h.

3.2 Index determination

In this subsection, we will determine the index of the differential-algebraic system in (10), since this tells us something about how strong the coupling between the equations is and, therefore, also something about numerical problems which may arise.

In the literature, various index concepts can be found. The probably most widely used are the differentiation index (d-index) introduced by Gear and Campbell [Gea88, Gea90, CG95] and the perturbation index (p-index) introduced by Hairer, Lubich and Roche [HLR89] (see also [HW96]). But there are also other concepts, e. g. the geometric index by Rheinboldt [Rhe84], the tractability index by Griepentrog and März [GM86], or the structural index by Pantelides [Pan88].

In this paper, the so-called “strangeness-index” (s-index) by Kunkel and Mehrmann [KM94, KM98] will be used for the analysis, since this concept is the most general one of all these index concepts. The strangeness-index can be seen as generalization of the differentiation index (see [KM94], [KM96b], and [KM01]) as well as of the perturbation index (see [KM96a]). Further, also over- and under-determined systems can be treated with this concept (see [KM01]).

A brief introduction to the strangeness-index of general (nonlinear) differential-algebraic equations can be found in Appendix B.

To determine the index of the nonlinear differential-algebraic system in (10), we proceed as described in Appendix B. Therefore, we first rewrite the system in the form

$$F(t, z, \dot{z}) = 0$$

with $t \in \mathbb{I}$, $z = \left[q_h^T, m_h^T, p_h^T\right]^T \in \mathbb{D}_z \subset \mathbb{R}^{\hat{n}}$, $\dot{z} = \left[\dot{q}_h^T, \dot{m}_h^T, \dot{p}_h^T\right]^T \in \mathbb{D}_{\dot{z}} \subset \mathbb{R}^{\hat{n}}$, $\hat{n} = n_q + n_m + n_p$, and $F \in \mathcal{C}(\mathbb{I} \times \mathbb{D}_z \times \mathbb{D}_{\dot{z}}, \mathbb{R}^{\hat{n}})$ given by

$$F = \begin{bmatrix} I_{n_q} & & \\ & 0 & \\ & & 0 \end{bmatrix} \begin{bmatrix} \dot{q}_h \\ \dot{m}_h \\ \dot{p}_h \end{bmatrix} - \begin{bmatrix} M^{-1}(\mathcal{G} + \mathcal{P}) \\ \mathcal{G}_5 \\ C_3^T u_h \end{bmatrix}.$$

Then we determine the Jacobians $E(t) := F_{\dot{z}}(t, z, \dot{z})$ and $A(t) := -F_z(t, z, \dot{z})$ as functions of t. Here, these matrices are given by

$$E = \begin{bmatrix} I_{n_q} & & \\ & 0 & \\ & & 0 \end{bmatrix} \quad \text{and} \quad A(t) = \begin{bmatrix} G(t) & 0 & C \\ 0 & R(t) & 0 \\ \tilde{C} & 0 & 0 \end{bmatrix},$$

where $G \in \mathcal{C}(\mathbb{I}, \mathbb{R}^{n_q \times n_q})$ and $R \in \mathcal{C}(\mathbb{I}, \mathbb{R}^{n_m \times n_m})$ denote the following Jacobians:

$$G = M^{-1}\frac{\partial \mathcal{G}}{\partial q_h}, \quad R = \frac{\partial \mathcal{G}_5}{\partial m_h}.$$

Moreover, $C \in \mathbb{R}^{n_q \times n_p}$ and $\tilde{C} \in \mathbb{R}^{n_p \times n_q}$ are given by

$$C = \begin{bmatrix} -\frac{1}{\rho_f} M_1^{-1} C_2 \\ 0 \\ 0 \\ 0 \end{bmatrix}, \quad \tilde{C} = \begin{bmatrix} C_3^T & 0 & 0 & 0 \end{bmatrix}$$

with C_2 and C_3 as defined before.

Let us now apply the theory from Appendix B to the nonlinear system, its Jacobians, and the corresponding inflated system. As shown in the appendix, the strangeness-index of a differential-algebraic equation can be determined by finding the smallest value $\hat{\mu} \in \mathbb{N}_0$ for which Hypothesis 2 from the appendix is fulfilled.
Therefore, we start with $\hat{\mu} = 0$ and check if Hypothesis 2 is satisfied in this case.

1. First, we have to determine $Z_2 \in \mathcal{C}(\mathbb{I}, \mathbb{R}^{\hat{n} \times \hat{a}})$ with maximal rank such that $Z_2^T \mathcal{M}_0 = 0$ holds. Since $\mathcal{M}_0$ is given by

$$\mathcal{M}_0 = \begin{bmatrix} I_{n_q} & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix},$$

it is clear that $\operatorname{rank}(\mathcal{M}_0) = n_q = \hat{n} - \hat{a}$ holds and we get the condition

$$\hat{a} = n_m + n_p. \tag{11}$$

Therefore, Z_2 is given by

$$Z_2 = \begin{bmatrix} 0 & 0 \\ I_{n_m} & 0 \\ 0 & I_{n_p} \end{bmatrix}.$$

Note that Z_2 is uniquely determined up to a transformation with a non-singular matrix (which does not have any influence on the rank).

2. Since the matrix A_2 is given by

$$\begin{aligned} A_2(t) \;=\; Z_2^T \mathcal{N}_0(t) \;&=\; \begin{bmatrix} 0 & I_{n_m} & 0 \\ 0 & 0 & I_{n_p} \end{bmatrix} \begin{bmatrix} G(t) & 0 & C \\ 0 & R(t) & 0 \\ \tilde{C} & 0 & 0 \end{bmatrix} \\ &=\; \begin{bmatrix} 0 & R(t) & 0 \\ \tilde{C} & 0 & 0 \end{bmatrix}, \end{aligned}$$

we get the condition $\hat{a} = \operatorname{rank}(\tilde{C}) + \operatorname{rank}(R)$ and further with (11)

$$\operatorname{rank}(\tilde{C}) = n_p, \quad \operatorname{rank}(R) = n_m, \tag{12}$$

which means that $\tilde{C}$ and R must both have full rank. Furthermore, $n_q \geq n_p$ must hold.

The matrix $T_2 \in \mathcal{C}(\mathbb{I}, \mathbb{R}^{\hat{n} \times \hat{d}})$ has to be chosen such that $Z_2^T \mathcal{N}_0 T_2 = 0$ holds. Therefore, we get

$$T_2 = \begin{bmatrix} 0 \\ 0 \\ I_{n_p} \end{bmatrix}.$$

3. It then follows that

$$\operatorname{rank}(ET_2) = \operatorname{rank}(\begin{bmatrix} 0 \\ 0 \\ 0 \end{bmatrix}) = 0 \stackrel{!}{=} \hat{d},$$

which leads to the contradiction $\hat{n} = \hat{d} + \hat{a} = n_m + n_p < \hat{n}$, since n_q is greater than zero. (Otherwise all the equations for the mean velocity, the turbulent kinetic energy, the energy dissipation rate, and the number density function would not occur in the system.)

Therefore, the strangeness-index of the system is greater than zero.

In the next step, we try out if Hypothesis 2 holds for $\hat{\mu} = 1$:

1. Again, we first determine a matrix $Z_2 \in \mathcal{C}(\mathbb{I}, \mathbb{R}^{2\hat{n} \times \hat{a}})$ which has maximal rank $\hat{a}$ (where $\hat{a}$ is defined by the rank of the kernel of $\mathcal{M}_1$) and fulfills

the condition $Z_2^T \mathcal{M}_1 = 0$. Since $\mathcal{M}_1 \in \mathcal{C}(\mathbb{I}, \mathbb{R}^{2\hat{n}\times 2\hat{n}})$ is given by

$$\mathcal{M}_1(t) = \left[\begin{array}{ccc|ccc} I_{n_q} & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ \hline -G(t) & 0 & -C & I_{n_q} & 0 & 0 \\ 0 & -R(t) & 0 & 0 & 0 & 0 \\ -\tilde{C} & 0 & 0 & 0 & 0 & 0 \end{array}\right],$$

we get the condition $\operatorname{rank}(\mathcal{M}_1) = 2n_q + \operatorname{rank}(R) \overset{!}{=} 2\hat{n} - \hat{a}$. Therefore, it follows that

$$\hat{a} = 2n_m + 2n_p - \operatorname{rank}(R) \tag{13}$$

and Z_2 is given by

$$Z_2^T(t) = \left[\begin{array}{ccc|ccc} 0 & I_{n_m} & 0 & 0 & 0 & 0 \\ 0 & 0 & I_{n_p} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & R^+(t) & 0 \\ \tilde{C} & 0 & 0 & 0 & 0 & I_{n_p} \end{array}\right],$$

where $R^+ \in \mathcal{C}(\mathbb{I}, \mathbb{R}^{n_m \times (n_m - \operatorname{rank}(R))})$ denotes the Co-range of R.

2. Then, we can determine $A_2 \in \mathcal{C}(\mathbb{I}, \mathbb{R}^{\hat{a}\times\hat{n}})$ by

$$A_2(t) = Z_2^T(t)\mathcal{N}_1(t) \begin{bmatrix} I_{\hat{n}} \\ 0 \end{bmatrix} = \begin{bmatrix} 0 & R(t) & 0 \\ \tilde{C} & 0 & 0 \\ 0 & R^+\dot{R}(t) & 0 \\ \tilde{C}G(t) & 0 & \tilde{C}C \end{bmatrix}.$$

Since the rank of A_2 has to be equal to $\hat{a}$, we get the following condition:

$$\hat{a} = \operatorname{rank}\left(\begin{bmatrix} \tilde{C} & 0 \\ \tilde{C}G & \tilde{C}C \end{bmatrix}\right) + \operatorname{rank}\left(\begin{bmatrix} R \\ R^+\dot{R} \end{bmatrix}\right).$$

With this, we get on the one hand

$$\hat{d} = \hat{n} - \hat{a} = n_q + n_m + n_p - \operatorname{rank}\left(\begin{bmatrix} \tilde{C} & 0 \\ \tilde{C}G & \tilde{C}C \end{bmatrix}\right) - \operatorname{rank}\left(\begin{bmatrix} R \\ R^+\dot{R} \end{bmatrix}\right),$$

whereas, on the other hand, Eq. (13) gives

$$\hat{d} = \hat{n} - \hat{a} = n_q - n_m - n_p + \operatorname{rank}(R).$$

By equating these two expressions and using the fact that

$$\begin{aligned} \operatorname{rank}\left(\begin{bmatrix} \tilde{C} & 0 \\ \tilde{C}G & \tilde{C}C \end{bmatrix}\right) &\leq n_p + \min(n_p, n_q), \\ \operatorname{rank}(R) &\leq n_m, \\ \operatorname{rank}\left(\begin{bmatrix} R \\ R^+\dot{R} \end{bmatrix}\right) &\leq n_m, \end{aligned}$$

this leads to the following conditions:

$$n_q \geq n_p, \quad \operatorname{rank}(R) = n_m, \quad \operatorname{rank}(\tilde{C}) = n_p, \quad \operatorname{rank}(\tilde{C}C) = n_p. \tag{14}$$

Therefore, it follows that the matrix R is nonsingular, and the third row in the matrix A_2 does not occur. Furthermore, $\hat{d}$ and $\hat{a}$ are given by $\hat{d} = n_q - n_p$ and $\hat{a} = 2n_p + n_m$.
Since the condition $A_2 T_2 = 0$ must hold, we get for $T_2 \in \mathcal{C}(\mathbb{I}, \mathbb{R}^{\hat{n} \times (n_q - n_p)})$:

$$T_2(t) = \begin{bmatrix} \tilde{C}^- \\ 0 \\ -(\tilde{C}C)^{-1}\tilde{C}G(t)\tilde{C}^- \end{bmatrix},$$

where $\tilde{C}^-$ denotes the kernel of the matrix $\tilde{C}$.

3. Furthermore, since $\operatorname{rank}(\tilde{C}) = n_p$ due to (14), it follows that

$$\operatorname{rank}(ET_2) = \operatorname{rank}(\tilde{C}^-) = n_q - n_p = \hat{d}$$

such that we can choose $Z_1 \in \mathcal{C}(\mathbb{I}, \mathbb{R}^{\hat{n} \times \hat{d}})$ as

$$Z_1^T = \begin{bmatrix} I_{n_q} & 0 & 0 \end{bmatrix},$$

yielding that $Z_1^T E(t) T_2 = \begin{bmatrix} \tilde{C}^- \end{bmatrix}$ has constant rank $\hat{d}$.

This shows that the strangeness-index of the differential-algebraic system in (10) corresponding to the partial differential equations, which describes the dynamical processes in the stirred tank, is equal to one if we choose the spatial discretization in such a way that the conditions in (14) are satisfied.

These conditions are easily fulfilled by using a sensible discretization. First of all, one can say that every discretization scheme should satisfy the condition $n_q \geq n_p$, since otherwise there would be more discretization points for the pressure than for the velocity, the turbulent kinetic energy, the energy dissipation rate, and the number density function together. Furthermore, in order to fulfill the condition that R is nonsingular, we only have to choose a suitable quadrature formula. In addition, the conditions $\operatorname{rank}(\tilde{C}) = n_p$ and $\operatorname{rank}(\tilde{C}C) = n_p$ are satisfied if the additive constant for the pressure is fixed and the continuity equation is discretized appropriately. This is due to the fact that in the Navier-Stokes equations, the pressure is only determined up to an additive constant, since only the gradient of the pressure occurs in the equations (see e. g. [Wes01]).

Since the rank conditions for $\tilde{C}$ and $\tilde{C}C$ already occur in the Navier-Stokes equations themselves, the considered system in (10) has the same strangeness-index as the Navier-Stokes equations. This means that the index of the Navier-Stokes equations was not increased by the coupling with the population balance equation. This observation can be summarized by the following corollary:

Corollary 1 *All solvers that are adapted to the solution of the Navier-Stokes equations are also suited for solving the discussed coupled system.*

Since the differential-algebraic system derived by the spatial discretization of the Navier-Stokes equations has strangeness-index one, it is hard to treat this system in its original form. When solving such a system as it appears originally, one can get into difficulties due to the mixing of differential and algebraic components. Therefore, it is useful to first remove this "strangeness" before solving the differential-algebraic system. Many Navier-Stokes solution techniques carry out such an index reduction, although this is often not mentioned explicitely [Wei96].

Among such typical solution methods are the so-called "penalty method", the pressure correction methods (also known as operator splitting methods), or the so-called "stream function-vorticity-pressure method" (see e. g. [Wes01], [Wei97]).

Within our project, the solver FEATFLOW [TB98] is used for the simulation of the Navier-Stokes equations. This method uses a finite element approach for the spatial discretization and a discrete projection scheme for the decoupling of pressure and velocity. In [Wei96], it has been shown that this kind of approach leads to a strangeness-free differential-algebraic system. Therefore, the approach used in FEATFLOW is suitable for the solution of the Navier-Stokes equations.

However, it must be reckoned with a order reduction in the time discretization [Wei96].

A Some models describing coalescence and dispersion processes

In this section we describe some possibilities how coalescence and dispersion processes can be modeled.

In the following we present the idea and the approach of the physically based model by Coulaloglou and Tavlarides [CT77]. Furthermore, we describe the differences to later introduced models by Tsouris and Tavlarides [TT94] and Ritter [Rit02].

In all these models, only one internal coordinate is used in the population balance equation in order to describe the properties of the particles. This internal coordinate is chosen to be the volume V of a particle or its diameter d. These two quantities are directly related to each other by the formula $V = c_V d^3$. (If we assume the drops to be spherical, we have $c_V = \frac{\pi}{6}$.)

As already mentioned before, the following models assume that coalescence and dispersion rates are space-independent. Additionally, the influence of the continuous phase vector is not taken into account in the models considered here.

Furthermore, the models underly the following assumptions [CT77]:

- The droplets are in a turbulent flow field which is locally isotropic. (Experimental investigations show that the flow field can be considered as locally isotropic for Re $\geq 10,000$ [CT77].) The droplet size d is within the following range: $\eta_{\text{macro}} \geq d \geq \eta_{\text{micro}}$, where η_{macro} and η_{micro} are the macroscale and microscale of turbulence, respectively.
- The energy spectrum function has a $-5/3$-dependency on the wave number, viscous effects are negligible, and the drop performs due to local pressure fluctuation.
- An oscillating deformed drop will break if the energy of the turbulent eddies exceeds the surface energy of the drop.

But as a first approach these models can also be used to describe the processes occurring in dispersions which do not fulfill all the assumptions above.

A.1 Coalescence

Coalescence is the confluence of fluid particles with each other or of a fluid particle with its mother phase. This means that smaller drops are formed together to larger ones.

The process of coalescence of two drops can be divided into three phases [Che91]:

- Collision of the drops,
- drainage of the film between the drops, and
- rupture of the film (after a critical thickness is reached) and confluence of the drops.

So, a necessary condition for coalescence of two drops is that they must remain in contact for a sufficiently long time so that the processes of film drainage, film rupture and coalescence may occur [CT77].

A.1.1 Coalescence rate

Coulaloglou and Tavlarides [CT77] model the coalescence rate R^{CT}_{coal} of two drops of volumes V and V' as a product of the collision frequency ϑ^{CT} and the coalescence efficiency λ^{CT}:

$$R^{CT}_{\text{coal}}(V,V') = \vartheta^{CT}(V,V')\lambda^{CT}(V,V').$$

They derive an expression for the collision frequency ϑ^{CT} by assuming that the mechanism of collision in a locally isotropic flow field is analogous to collisions between molecules as in the kinetic theory of gases. Hence they use the following formula:

$$\vartheta^{CT}(V,V') = \left(\frac{9\pi}{2}\right)^{1/3} (V^{2/3} + V'^{2/3})(\overline{u^2}(V) + \overline{u^2}(V'))^{1/2}, \tag{15}$$

where $\overline{u^2}(V)$ is the mean square fluctuation velocity of a drop of volume V, given by

$$\overline{u^2}(V) = \overline{c}_1\, \varepsilon_l^{2/3} V^{2/9}, \tag{16}$$

where ε_l is the local energy dissipation rate per unit mass and $\overline{c}_1$ is a dimensionless constant. In [CT77] it is assumed that ε_l is constant throughout the tank and given by the following relation:

$$\varepsilon_l \sim N^{*3} D_{\text{imp}}, \tag{17}$$

where N^* is the impeller speed (revolutions per time) and D_{imp} is the impeller diameter.

Inserting this into Eq. (15) yields

$$\vartheta^{CT}(V,V') = \overline{c}_2\, \varepsilon_l^{1/3} (V^{2/3} + V'^{2/3})(V^{2/9} + V'^{2/9})^{1/2}$$

with a constant $\overline{c}_2$.

According to Coulaloglou and Tavlarides [CT77], the coalescence efficiency λ^{CT} can be related to the physical phenomena which occur: The drops must be compressed for a sufficient time so that the film between them can rupture and the drops are able to coalesce. This means that the drops have to be in contact for a longer time than coalescence needs to take place. In [CT77] the following approach is used:

$$\lambda^{CT}(V,V') = e^{-\overline{t}^{CT}_{\text{coal}}/\overline{t}^{CT}_{\text{contact}}}, \tag{18}$$

where $\overline{t}^{CT}_{\text{coal}}$ and $\overline{t}^{CT}_{\text{contact}}$ are the averages of coalescence and contact time, respectively.

Coulaloglou and Tavlarides [CT77] estimate the coalescence time as the time required for film drainage between the drops. They use

$$\overline{t}^{CT}_{\text{coal}} \sim \frac{3}{16} \frac{\mu_c K}{\pi \sigma^2} \left(\frac{1}{h_1^2} - \frac{1}{h_0^2}\right) \left(\frac{dd'}{d+d'}\right)^2, \tag{19}$$

where K is the force compressing the drops, given by

$$K \sim \rho_c \overline{u^2} \left(\frac{dd'}{d+d'}\right)^2, \tag{20}$$

with $\overline{u^2} \sim \varepsilon_l^{2/3}(d+d')^{2/3}$ due to Eq. (16). Here, μ_c and ρ_c are viscosity and density of the continuous phase, respectively, and σ is the interfacial tension.

(The model assumes that the drops are located in a fluid eddy of size $d+d'$.) The two parameters h_0 and h_1 are the film thicknesses at initial contact of the drops and the one at the time when spontaneous film rupture can occur. Inserting the formulas for $\overline{u^2}$ and K into Eq. (19) leads to

$$\bar{t}^{CT}_{\text{coal}} \sim \frac{\mu_c \rho_c \varepsilon_l^{2/3} (d+d')^{2/3}}{\sigma^2} \left(\frac{1}{h_1^2} - \frac{1}{h_0^2} \right) \left(\frac{dd'}{d+d'} \right)^4 .$$

Coulaloglou and Tavlarides estimate the contact time $\bar{t}^{CT}_{\text{contact}}$ as the time two drops of size d and d' will stay together in a turbulent flow:

$$\bar{t}^{CT}_{\text{contact}} \sim \frac{(d+d')^{2/3}}{\varepsilon_l^{1/3}}.$$

Altogether the coalescence efficiency can be written as

$$\lambda^{CT}(V,V') = \exp\left(-\bar{c}_3 \frac{\mu_c \rho_c \varepsilon_l}{\sigma^2} \left(\frac{V^{1/3} V'^{1/3}}{V^{1/3} + V'^{1/3}} \right)^4 \right), \tag{21}$$

since the film thicknesses h_0 and h_1 are assumed to be constant.

Therefore, the coalescence rate R^{CT}_{coal} by Coulaloglou and Tavlarides [CT77] is given by

$$\begin{aligned} R^{CT}_{\text{coal}}(V,V') &= \overline{C}_1 \left(V^{2/3} + V'^{2/3} \right) \left(V^{2/9} + V'^{2/9} \right)^{1/2} \varepsilon_l^{1/3} \\ & \quad \exp\left(-\overline{C}_2 \frac{\mu_c \rho_c \varepsilon_l}{\sigma^2} \left(\frac{V^{1/3} V'^{1/3}}{V^{1/3} + V'^{1/3}} \right)^4 \right), \end{aligned}$$

where $\overline{C}_1$ and $\overline{C}_2$ are adaption parameters.

After comparing numerical and experimental results, Coulaloglou and Tavlarides found a damping of turbulence depending on the volume fraction χ of the dispersed phase. To take this into account, they used a different formula for the velocity in a dispersion:

$$\overline{u_\chi^2} = (1+\chi)^{-2} \overline{u^2}. \tag{22}$$

Inserting this into the Formulas (15) and (20), then leads to

$$\begin{aligned} \tilde{R}^{CT,\chi}_{\text{coal}}(V,V') &= \overline{C}_1 \frac{\varepsilon_l^{1/3}}{1+\chi} \left(V^{2/3} + V'^{2/3} \right) \left(V^{2/9} + V'^{2/9} \right)^{1/2} \\ & \quad \exp\left(-\overline{C}_2 \frac{\mu_c \rho_c \varepsilon_l}{\sigma^2 (1+\chi)^2} \left(\frac{V^{1/3} V'^{1/3}}{V^{1/3} + V'^{1/3}} \right)^4 \right). \end{aligned}$$

But Coulaloglou and Tavlarides [CT77] derived a slightly different final form for their coalescence rate $R^{CT,\chi}_{\text{coal}}$: Instead of the term $(1+\chi)^2$ in the denominator of the exponential function, they have the term $(1+\chi)^3$, but besides this, there are no further differences.

Tsouris and Tavlarides [TT94] use basically the same approach as Coulaloglou and Tavlarides [CT77] to model the coalescence rate, i. e. they also express the coalescence rate by the product of collision frequency and coalescence efficiency. Furthermore, they also model the collision frequency by assuming that the drops in a turbulent flow behave like gas molecules, but use the following (slightly different) formula:

$$\vartheta^{TT}(d,d') = \frac{\pi}{4}\left(d+d'\right)^2\left(\overline{u^2}(d)+\overline{u^2}(d')\right)^{1/2} \quad \text{with} \quad \overline{u^2}(d) = 1.07\varepsilon_l^{2/3}d^{2/3}, \tag{23}$$

where the constant ε_l is given by Eq. (17) as proposed in [CT77]. As approach for the coalescence efficiency, they also use Eq. (18).

The only main difference between the two models lies in the formulas for coalescence and contact time. In [TT94], the averaged coalescence time $\bar{t}^{TT}_{\text{coal}}$ between two drops with diameters d and d' is given by

$$\bar{t}^{TT}_{\text{coal}} = \overline{c}_4\frac{6\pi\mu_c\overline{c}_5}{\rho_c\varepsilon_l^{2/3}(d+d')^{2/3}},$$

where μ_c and ρ_c are the viscosity and density of the continuous phase, respectively. Tsouris and Tavlarides use the value 3.44 for the constant $\overline{c}_4$. The parameter $\overline{c}_5$ is given by

$$\overline{c}_5 = 1.872\ln\left[\frac{h_0^{1/2}+1.378\overline{c}_6}{h_1^{1/2}+1.378\overline{c}_6}\right] + 0.127\ln\left[\frac{h_0^{1/2}+0.312\overline{c}_6}{h_1^{1/2}+0.312\overline{c}_6}\right],$$

where h_0 and h_1 are the film thicknesses at initial contact of the drops and at the time when spontaneous film rupture can occur, respectively. The parameter $\overline{c}_6$ is given by

$$\overline{c}_6 = \frac{\mu_c}{\mu_d}\left(\frac{dd'}{2(d+d')}\right)^{1/2},$$

where μ_d is the viscosity of the dispersed phase. For the contact time $\bar{t}^{TT}_{\text{contact}}$ of two drops with diameters d and d' they use the following formula:

$$\bar{t}^{TT}_{\text{contact}} = \frac{(D^2_{\text{tank}}H_{\text{tank}})^{1/3}}{31.25N^*D_{\text{imp}}},$$

where D_{tank} is the vessel diameter, H_{tank} is the tank height, N^* is the impeller speed, and D_{imp} is the impeller diameter.

Ritter [Rit02] uses a kind of mixture of the two models introduced above. For the collision frequency he uses Eq. (23) by Tsouris and Tavlarides, whereas he models the coalescence efficiency by Eq. (21) given by Coulaloglou and Tavlarides. Further, he includes a damping factor DF, depending on the volume fraction χ, given by

$$DF(\chi) = \left[1 + 2.5\,\chi\,\frac{\mu_d + 0.4\mu_c}{\mu_d + \mu_c}\right]^2, \tag{24}$$

in both functions, the collision frequency as well as the coalescence efficiency. Here, as before, μ_d and μ_c denote the dynamic viscosity of the dispersed and the continuous phase, respectively. Note that this damping factor was already used by Tsouris and Tavlarides to take into account the turbulence damping of the dispersed phase when modeling the dispersion process. Altogether, the coalescence rate used by Ritter is given by

$$\begin{aligned} R^R_{\mathrm{coal}}(d,d') &= \overline{C}_3 \frac{\varepsilon_l^{1/3}}{DF(\chi)^{1/2}} (d+d')^2 \left(d^{2/3} + d'^{2/3}\right)^{1/2} \\ &\quad \exp\left(-\overline{C}_4 \frac{\mu_c \rho_c \varepsilon_l}{\sigma^2 DF(\chi)^{3/2}} \left(\frac{dd'}{d+d'}\right)^4\right). \end{aligned}$$

A.2 Dispersion

Dispersion means the breakup of larger particles into smaller ones. This can be originated by the collision of individuals with each other or with the stirrer or by shear stresses caused by the streaming.

Deformation and breakage of a drop in a turbulent flow depends on many parameters, e. g., drop size, density, interfacial surface tension, viscosity of both phases, holdup fraction, local flow, and local energy dissipation (see [CT77]).

A.2.1 Dispersion rate

Again, we first consider the approach by Coulaloglou and Tavlarides [CT77], which is based on the nature of dispersion.

They use the following approach for the dispersion (or breakage) rate of a droplet of volume V:

$$R^{CT}_{\mathrm{disp}}(V) = \frac{1}{t_b}\frac{\Delta N(V)}{N(V)},$$

where t_b is the breakage time and $\frac{\Delta N(V)}{N(V)}$ is the fraction of drops breaking. This fraction is assumed to be proportional to the fraction of turbulent eddies colliding with the drop and having a turbulent kinetic energy greater than the surface energy of the drop:

$$\frac{\Delta N(d)}{N(d)} = e^{-E_\sigma/\overline{E}},$$

where $E_\sigma(d)$ is the surface energy of a drop with diameter d and $\overline{E}(d)$ is the mean turbulent kinetic energy of an eddy. For the surface energy E_σ the formula

$$E_\sigma(d) = \tilde{c}_1\, \sigma d^2$$

is used, while the mean turbulent kinetic energy $\overline{E}$ is given by

$$\overline{E}(d) = \tilde{c}_2\, \rho_d d^3 \overline{u^2}(d), \tag{25}$$

where σ is the interfacial tension and ρ_d is the density of the dispersed phase. The mean square of the relative velocity $\overline{u^2}(d)$ between two points separated by a distance d in the inertial subrange is again given by

$$\overline{u^2}(d) = \tilde{c}_3\, \varepsilon_l^{2/3} d^{2/3},$$

where for ε_l again Eq. (17) can be used.

The breakage time t_b is estimated by assuming that the motion of the centers of mass of the daughter drops is similar to the relative motion of two lumps of fluid in a turbulent flow field:

$$t_b \sim d^{2/3} \varepsilon_l^{-1/3}.$$

Altogether the dispersion rate by Coulaloglou and Tavlarides [CT77] is given by

$$R_{\text{disp}}^{CT}(V) = \tilde{C}_1 V^{-2/9} \varepsilon_l^{1/3} \exp\left(-\frac{\tilde{C}_2 \sigma}{\rho_d \varepsilon_l^{2/3} V^{5/9}} \right)$$

with two dimensionless constants $\tilde{C}_1$ and $\tilde{C}_2$. They can be used as adaption parameters.

Taking again the turbulence damping due to the volume fraction χ of the dispersed phase into account, their dispersion rate becomes

$$R_{\text{disp}}^{CT,\chi}(V) = \tilde{C}_1 \frac{\varepsilon_l^{1/3}}{1+\chi} V^{-2/9} \exp\left(-\frac{\tilde{C}_2 \sigma (1+\chi)^2}{\rho_d \varepsilon_l^{2/3} V^{5/9}} \right).$$

Again, we get a slightly different formula if we just substitute $\overline{u^2}$ by $\overline{u_\chi^2}$ in Eq. (25), namely:

$$\tilde{R}_{\text{disp}}^{CT,\chi}(V) = \tilde{C}_1 \varepsilon_l^{1/3} V^{-2/9} \exp\left(-\frac{\tilde{C}_2 \sigma (1+\chi)^2}{\rho_d \varepsilon_l^{2/3} V^{5/9}} \right).$$

Let us now have a closer look at the model of Tsouris and Tavlarides [TT94]. They use the approach of Prince and Blanch [PB90] to model the dispersion (or breakage) rate. There, the dispersion rate R_{disp}^{TT} of a drop with diameter

d is given by the product of eddy-drop-collision frequency ϑ^{TT} and breakage efficiency B^{TT}:

$$R_{\text{disp}}^{TT}(d) = \vartheta^{TT}(d)B^{TT}(d).$$

In their model, the eddy-drop-collision frequency is modeled (like the drop-drop collision frequency in the model of Coulaloglou and Tavlarides [CT77]) by assuming that the eddies and drops move like ideal gas molecules. Therefore, their collision process can be described with the help of the kinetic theory of gases.

Accordingly, Tsouris and Tavlarides state for the collision frequency between drops of size d and eddies of a size range that can break these drops:

$$\vartheta^{TT}(d) = \int \frac{\pi}{4}(d_e + d)^2(u_e^2 + u_d^2)^{1/2} dn_e, \tag{26}$$

where d_e is the size of an eddy that can break a drop with diameter d and dn_e is the number of eddies of size between d_e and $d_e + \delta d_e$. Eddy velocity u_e and drop velocity u_d are given by

$$u_e^2 = 8.2\left(\frac{\varepsilon_l}{\kappa_e}\right)^{2/3} \quad \text{and} \quad u_d^2 = 1.07\varepsilon_l^{2/3}d^{2/3},$$

where ε_l is chosen as before and $\kappa_e = \frac{2}{d_e}$ is the wave number of an eddy with diameter d_e.

Inserting these two formulas into Eq. (26) (and using $d_e = \frac{2}{\kappa_e}$) gives

$$\vartheta^{TT}(d) = \int \frac{\pi}{4}\left(\frac{2}{\kappa_e} + d\right)^2 \varepsilon_l^{1/3}\left[8.2\kappa_e^{-2/3} + 1.07d^{2/3}\right]^{1/2} dn_e.$$

Tsouris and Tavlarides substitute the integration variable n_e by the eddy wave number κ_e by using the following differential equation for the number of eddies per unit mass of the fluid $n_{e,m}$:

$$\frac{dn_{e,m}(\kappa_e)}{d\kappa_e} = 0.1\frac{\kappa_e^2}{\rho_f},$$

where ρ_f is the density of the fluid. Hence, the collision frequency by Tsouris and Tavlarides is given by

$$\vartheta^{TT}(d) = \tilde{c}_4\varepsilon_l^{1/3}\int_{2/d}^{2/d_{e,\min}}\left(\frac{2}{\kappa_e} + d\right)^2\left[8.2\kappa_e^{-2/3} + 1.07d^{2/3}\right]^{1/2}\kappa_e^2 d\kappa_e.$$

Here, the integral term represents the total number of eddies that are able to break drops with diameter d. The upper limit of the integration is the (large) wave number that corresponds to the eddy size $d_{e,\min}$. This eddy size $d_{e,\min}$ can be taken arbitrarily as half of the critical drop size, since eddies of size less than the critical drop diameter are not able to break any drop in the system because they do not have enough kinetic energy. In [TT94] the constant $\tilde{c}_4$ includes a turbulence damping factor $DF(\chi)$ due to the dispersion as well as the fraction of the volume of the impeller region V_i to the total volume V_t of the tank:

$$\tilde{c}_4 = \frac{V_i}{V_t}\frac{0.1\pi}{4}DF(\chi),$$

where the damping factor $DF(\chi)$ is given by Eq. (24).

For the breakage efficiency Tsouris and Tavlarides use the approach to describe it by an exponential function (cf. Coulaloglou and Tavlarides [CT77] or Prince and Blanch [PB90]). This approach accounts for the different forces acting on the drop. (A drop breaks if its surface tension is exceeded by the energy of a colliding eddy.) Therefore, the breakage efficiency can be described by

$$B^{TT} = \exp\left(-\frac{\overline{E}_\sigma}{\tilde{c}_5\overline{E}_{\text{eddy}}}\right),$$

where $\overline{E}_\sigma$ is the average energy required for drop breakage, $\overline{E}_{\text{eddy}}$ is the average energy of an eddy, and $\tilde{c}_5$ is a constant. The average energy required for drop breakage is assumed to be the arithmetic mean of the minimal and maximal energy required for forming two daughter drops. The minimal energy is given by the energy needed to form the smallest and largest possible daughter drop, the maximal energy by the one needed to form two equal-size drops. Therefore, we get

$$\overline{E}_\sigma = \frac{1}{2}\Big\{\underbrace{2\pi\sigma\left(\frac{d}{2^{1/3}}\right)^2}_{\text{maximal energy } E_{\sigma,\max}} + \underbrace{\pi\sigma d_{\max}^2 + \pi\sigma d_{\min}^2}_{\text{minimal energy } E_{\sigma,\min}}\Big\} - \underbrace{\pi\sigma d^2}_{\text{energy of breaking drop}},$$

where σ is the interfacial tension and $d_{\max}$ and $d_{\min}$ are the diameters of the largest and smallest possible drops which can be formed by dispersion of a drop with diameter d. (Note that, due to conservation of mass, the formula $d^3 = d_{\max}^3 + d_{\min}^3$ must hold.) The average energy of an eddy is given by

$$\overline{E}_{\text{eddy}} = 0.43\pi\rho_f\varepsilon_l^{2/3}d_e^{11/3} \overset{d_e=\frac{2}{\kappa_e}}{=} 5.47\pi\rho_f\varepsilon_l^{2/3}\kappa_e^{-11/3}.$$

Altogether the dispersion rate by Tsouris and Tavlarides [TT94] is given by

$$R_{\text{disp}}^{TT}(d) = \tilde{C}_3 DF(\chi)\varepsilon_l^{1/3}\int_{2/d}^{2/d_{e,\min}}\left(\frac{2}{\kappa_e}+d\right)^2\left[8.2\kappa_e^{-2/3}+1.07d^{2/3}\right]^{1/2} \exp\left(-\frac{\overline{E}_\sigma}{\tilde{C}_4\overline{E}_{\text{eddy}}}\right)\kappa_e^2 d\kappa_e, \quad (27)$$

where $DF(\chi)$ is the (above mentioned) damping factor dependent on the volume fraction χ of the dispersed phase. Tsouris and Tavlarides use the following values for the constants $\tilde{C}_3$ and $\tilde{C}_4$:

$$\tilde{C}_3 = 0.0118 \quad \text{and} \quad \tilde{C}_4 = 1.3.$$

The dispersion rate by Ritter [Rit02] is quite similar to that one by Tsouris and Tavlarides. The only difference is that Ritter puts the damping factor into the denominator and takes the square root of it. In [Rit02] the constants $\tilde{C}_3$ and $\tilde{C}_4$ are given by

$$\tilde{C}_3 = 0.1\frac{\pi}{4} \approx 0.0785 \quad \text{and} \quad \tilde{C}_4 = 1.$$

A.2.2 Distribution of daughter drops

To be able to model the source term due to dispersion, a distribution of daughter drops is needed. The distribution of daughter drops is a probability density function which states the probability that the dispersion of a drop of diameter d' (or volume V') leads to a daughter drop of diameter d (or volume V). The distribution of daughter drops (or breakage distribution function) can be modeled in different ways. Many different approaches can be found in the literature (see e. g. [CT77], [TT94] and [Rit02]).

Coulaloglou and Tavlarides [CT77] assume that the distribution of daughter drops takes the form

$$\gamma^{CT}(V,V') = \frac{2.4}{V'} \exp\left(-4.5\frac{(2V-V')^2}{V'^2}\right),$$

i. e. they use a normal distribution. Here, the variance is chosen such that more than 99.6 percent of the droplets formed lie within the volume range 0 to V'. Note that this function can be rewritten in the following form:

$$\gamma^{CT}(V,V') = \frac{1}{\frac{V'}{6}\sqrt{2\pi}} \exp\left(\frac{(V-\frac{1}{2}V')^2}{2\left(\frac{V'}{6}\right)^2}\right). \tag{28}$$

Ritter [Rit02] also uses a normal distribution, given by the following formula:

$$\gamma^{R}(V,V') = \frac{1}{\frac{V'}{10}\sqrt{2\pi}} \exp\left(-\frac{\left(V-\frac{V'}{2}\right)^2}{2\left(\frac{V'}{10}\right)^2}\right).$$

Comparison with Eq. (28) shows that the normal distribution used by Ritter has a smaller variance than the one used by Coulaloglou and Tavlarides.

It can be shown that the energy requirement for a breakage into two equal-size drops is greater than the energy requirement for a breakage into a small and a large drop [TT94]. The use of a normal distribution, which is unimodal and has a higher probability density in the middle, does not take this into account. Therefore, it is better to use a bimodal distribution with higher probability at the ends than in the middle.

Tsouris and Tavlarides [TT94] use the following function for the distribution of daughter drops:

$$\gamma^{TT}(d, d') = \frac{E_{\min}(d) + E_{\max}(d) - E(d)}{\int_0^{d'} \left[E_{\min}(\delta) + E_{\max}(\delta) - E(\delta)\right] d\delta},$$

where $E_{\min}(d)$ and $E_{\max}(d)$ are the minimal and maximal energy which is needed for dispersion of a drop with diameter d into two daughter drops, and $E(d)$ is the energy of a dispersing drop of diameter d.

However, they only give concrete formulas for these energies for the discrete case. Therefore, we will use the above introduced formulas to model these energy terms:

$$\begin{aligned} E_{\min}(d) &:= E_{\sigma,\min}(d) = \pi\sigma d_{\max}^2 + \pi\sigma d_{\min}^2 \quad \text{with } d_{\max}^3 + d_{\min}^3 = d^3, \\ E_{\max}(d) &:= E_{\sigma,\max}(d) = 2\pi\sigma \left(\frac{d}{2^{1/3}}\right)^2, \\ E(d) &:= \pi\sigma d^2. \end{aligned}$$

In [Ger99] another bimodal distribution can be found:

$$\gamma_*^G(V, V') = V'(V' - V) \exp\left(-\hat{C}_1^4 \left(\left(\frac{V'}{V} - \frac{1}{2}\right)^4 - \left(\frac{1 - 2\hat{C}_2}{\sqrt{2}} \left(\frac{V'}{V} - \frac{1}{2}\right)\right)^2\right)\right)$$

with constants $\hat{C}_1 = 0.065$ and $\hat{C}_2 = 3.5$. This function has to be scaled such that it fulfills the normalization condition (4). This leads to:

$$\gamma^G(V, V') = \frac{\gamma_*^G(V, V')}{\int_0^{V_{\max}} \gamma_*^G(V, V') dV'}.$$

In general, for every distribution of daughter drops, the dependency on the internal coordinate can be changed from volume V to diameter d (and vice versa) by the following transformation:

$$\gamma(d, d') = 3c_V d^2 \gamma(V, V'),$$

where c_V is the shape factor between volume and diameter. (It holds $c_V = \frac{\pi}{6}$ for spherical drops.)

B How to determine the strangeness-index of a nonlinear differential-algebraic equation

In this section, we give a brief introduction how the strangeness-index of a nonlinear differential-algebraic equation is defined and how it can be determined. This introduction is a brief summary of parts of the theory presented in [KM98].

We consider a general differential-algebraic equation

$$F(t,z,\dot z)=0, \tag{29}$$

with $F\in\mathcal{C}(\mathbb{I}\times D_z\times D_{\dot z},\mathbb{R}^{\hat n})$, $\mathbb{I}\subset\mathbb{R}$ (compact) interval, $D_z,D_{\dot z}\subset\mathbb{R}^{\hat n}$ open.

The standard method for the solution of nonlinear problems is Newton's method. There, we first linearize around some given initial guess, then solve the so obtained linear problem and afterwards correct the initial guess.
This suggests to consider any linearization of (29) and apply the theory for linear systems to the resulting equations in order to determine the index of the nonlinear system given in (29).
But counter-examples show that for higher index differential-algebraic equations it makes no sense to consider linearizations of the original equation alone. We must also include derivatives of the equation. Thus, we define the so-called "inflated differential-algebraic equation", which consists of the original equation and its derivatives up to order $l\in\mathbb{N}_0$, by

$$\begin{aligned}
F_l(t,z,\dot z,\ldots,z^{(l+1)}) &= \begin{bmatrix} F(t,z,\dot z)\\ \frac{d}{dt}F(t,z,\dot z)\\ \vdots\\ (\frac{d}{dt})^l F(t,z,\dot z)\end{bmatrix}\\
&= \begin{bmatrix} F(t,z,\dot z)\\ F_t(t,z,\dot z)+F_z(t,z,\dot z)\dot z+F_{\dot z}(t,z,\dot z)\ddot z\\ \vdots\end{bmatrix} = 0
\end{aligned}$$

and denote its Jacobians by

$$\begin{aligned}
\mathcal{M}_l(t,z,\dot z,\ldots,z^{(l+1)}) &= F_{l;\dot z,\ldots,z^{(l+1)}}(t,z,\dot z,\ldots,z^{(l+1)}),\\
\mathcal{N}_l(t,z,\dot z,\ldots,z^{(l+1)}) &= -\left[F_{l;z}(t,z,\dot z,\ldots,z^{(l+1)}),0,\ldots,0\right].
\end{aligned}$$

If we define

$$E:=F_{\dot z}(t,z,\dot z)\quad\text{and}\quad A:=-F_z(t,z,\dot z),$$

the Jacobians $\mathcal{M}_l$ and $\mathcal{N}_l$ can be written as in the linear case (see [KM96b], for example):

$$\mathcal{M}_l = \begin{bmatrix} E & & & & \\ \dot E - A & E & & & \\ \ddot E - 2\dot A & 2\dot E - A & E & & \\ \vdots & & \ddots & \ddots & \\ E^{(l)}-lA^{(l-1)} & \ldots & & l\dot E - A & E \end{bmatrix},$$

$$\mathcal{N}_l = \begin{bmatrix} A & 0 & \ldots & 0\\ \dot A & 0 & \ldots & 0\\ \ddot A & 0 & \ldots & 0\\ \vdots & \vdots & & \vdots\\ A^{(l)} & 0 & \ldots & 0\end{bmatrix}.$$

The following hypothesis shows an approach to determine the invariants of a nonlinear differential-algebraic equation.

Hypothesis 2 *There exist integers $\hat{\mu}$, $\hat{a}$, and $\hat{d}$ such that for all values $(t, z, \dot{z}, \ldots, z^{(\hat{\mu}+1)}) \in L$, where L associated with F is given by*

$$L = \{(t, z, \dot{z}, \ldots, z^{(\hat{\mu}+1)}) \in \mathbb{I} \times \mathbb{R}^{\hat{n}} \times \mathbb{R}^{\hat{n}} \times \cdots \times \mathbb{R}^{\hat{n}} \mid F_{\hat{\mu}}(t, z, \dot{z}, \ldots, z^{(\hat{\mu}+1)}) = 0\}$$

and $F_{\hat{\mu}}(t, z, \dot{z}, \ldots, z^{(\hat{\mu}+1)}) = 0$ is considered as an algebraic equation, the following properties are satisfied.

1. *It holds that*

$$\operatorname{rank}(\mathcal{M}_{\hat{\mu}}(t, z, \dot{z}, \ldots, z^{(l+1)})) = (\hat{\mu} + 1)\hat{n} - \hat{a},$$

such that there exists a matrix function Z_2 of size $(\hat{\mu} + 1)\hat{n} \times \hat{a}$ and with maximal rank which is smooth on L and satisfies there $Z_2^T \mathcal{M}_{\hat{\mu}} = 0$.

2. *It holds that*

$$\operatorname{rank}(A_2(t, z, \dot{z}, \ldots, z^{(l+1)})) = \hat{a} \quad \textit{with} \quad A_2 = Z_2^T \mathcal{N}_{\hat{\mu}}[I_{\hat{n}}\ 0\ \ldots\ 0]^T,$$

such that there exists a matrix function T_2 of size $n \times \hat{d}$, $\hat{d} = n - \hat{a}$, and with maximal rank which is smooth on L and satisfies there $A_2 T_2 = 0$.

3. *It holds that*

$$\operatorname{rank}(F_{\dot{z}}(t, z, \dot{z}) T_2(t, z, \dot{z}, \ldots, z^{(l+1)})) = \hat{d},$$

such that there exists a matrix function Z_1 of size $\hat{n} \times \hat{d}$ which is smooth on L and satisfies there $\operatorname{rank}(Z_1^T E) = \hat{d}$ (such that $Z_1^T E T_2$ has full rank $\hat{d}$ on L).

By means of this hypothesis we can define the strangeness-index:

Definition 3 *The strangeness-index of the nonlinear differential-algebraic equation given in (29) is the smallest integer $\hat{\mu}$ for which Hypothesis 2 is satisfied.*

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