Modeling drop breakage using the full energy spectrum and a specific realization of turbulence anisotropy

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Abstract
The assumption of homogeneous isotropic turbulence when modeling drop breakage in industrially relevant geometries is questionable. We describe the development of an anisotropic breakage model, where the anisotropy is introduced via the inclusion of a perturbed turbulence spectrum. The selection of the perturbed spectrum is itself motivated by our previous large-eddy simulations of high-pressure homogenizers. The model redistributes energy from small to large scales, and assumes that the anisotropic part of the Reynolds stresses is confined to the energy-containing range. The second-order structure function arising from the perturbed spectrum is used in the standard framework of Coulaloglou and Tavlarides to calculate breakage frequency. While the base model exhibits non-monotonic behavior (by predicting a maximum value for a certain drop size), the effect of anisotropy is shown to increase breakage frequency in length scales larger than this peak, thereby reducing non-monotonicity. This effect is more pronounced for small turbulence Reynolds numbers.

KEYWORDS
mathematical modeling, particulate flows, process, simulation, turbulence

1 | INTRODUCTION

Immiscible liquid–liquid systems are commonly encountered in chemical, petroleum, fast-moving consumer goods and pharmaceutical industries. Process design, scale-up and optimization still rely on time-consuming and costly experimental programs, motivating in-silico evaluation of emulsification processes via simulation techniques. The drop size distribution (DSD) in such systems is important because it determines critical features of the final product and affects the process dynamics through rheology. The evolution of the DSD is often modeled through a population balance equation (PBE), which is a continuity statement for the population of drops. Several methods have been developed to solve the PBE, such as the classes method, the Monte Carlo method, and the methods of moments; particularly important are the quadrature-based moment methods, since they are a computationally tractable approach to solve the PBE coupled to CFD. These methods are suitable when the distribution of the dispersed phase is simple (e.g., monomodal or bimodal), and the main interest is on its average properties rather than on the complete distribution. For more complex distributions, a large number of moments is required which may impact the stability of the solution when higher-order schemes are employed.

A key challenge that applies to all the discussed methods lies in the submodels describing drop breakage and coalescence. Many competing models have been published over many years, and a detailed overview of them is given in the review papers of Lasheras et al, Liao and Lucas, Solsvik et al, Sajjadi et al, Abidin et al, and Falzone et al. In this study, we focus on drop breakage frequency in turbulent flow. A major disagreement between the different
models concerns their monotonic or non-monotonic behavior with increasing drop size. Early models and their extensions\textsuperscript{21-24} predict a maximum breakage frequency value for certain drop size, while later models\textsuperscript{25-28} impose monotonicity based on available experimental evidence,\textsuperscript{25} arguing that a continuously rising breakage frequency with an increasing drop size is physically more plausible. However, this argument is questionable as recent experimental data\textsuperscript{24} have shown non-monotonic behavior in the breakage frequency itself.

Drop breakage in turbulent flows is generally modeled in terms of Kolmogorov’s second similarity hypothesis\textsuperscript{29} and the second-order longitudinal structure function ($\langle \Delta u^2 \rangle$) for the inertial subrange of homogeneous isotropic turbulence (HIT).\textsuperscript{30} Following the functional form of Coulaloglou and Tavlarides,\textsuperscript{21} a general form of drop breakage frequency may be written as\textsuperscript{24}

$$b(\xi) \propto t_b^{-1}(\xi)P_b(\xi),$$

(1)

where $P_b$ is the breakage probability, $\xi$ is the drop diameter, and the breakage time is given by

$$t_b(\xi) \propto \frac{\xi}{\sqrt{\langle \Delta u^2 \rangle}}.$$  

(2)

The underlying assumption is that the motion of the daughter drops is essentially that of turbulent structures in the flow field with the same length scale\textsuperscript{21}; thus the time needed for drop breakage to occur is assumed to be proportional to the turbulent time scale at the length scale of the drop. According to the authors,\textsuperscript{21} the constant of proportionality is determined by experiment. The longitudinal (as opposed to transverse) structure function is used because, following the Kolmogorov–Hinze theory of emulsification,\textsuperscript{31,32} the dynamic pressure fluctuations corresponding to the turbulent normal stresses are considered responsible for drop breakage.

The most quoted model for breakage frequency\textsuperscript{18,33} is the one of Coulaloglou and Tavlarides\textsuperscript{21} and its extensions\textsuperscript{22-24}; the model is written as

$$b(\xi) = c_{1b}^{1/3} \exp \left( - \frac{c_{2b}b}{\rho_c \langle \Delta u^2 \rangle^{1/3}} - \frac{c_{3b}b}{\rho_d \langle \Delta u^2 \rangle^{1/3}} \right).$$

(3)

$\varepsilon$ is the dissipation rate of the turbulent kinetic energy, $\sigma$ is the interfacial tension, $\mu_d$ is the dispersed phase dynamic viscosity, $\rho_c$ and $\rho_d$ are the continuous and the dispersed phase densities, respectively, and $c_{1b}$, $b - c_{3b}$ are model parameters. Note that Equation (3) was proposed by Chen et al\textsuperscript{34} as an extension of the original model, to include the effect of the viscous stresses in the $P_b$ term; this model formulation is used throughout the present study.

The breakage time in Equation (3) may be obtained by substituting $\langle \Delta u^2 \rangle$ in Equation (2) with $\beta(\xi)\sigma^{2/3}$.\textsuperscript{29} The constant $\beta$, linking $\langle \Delta u^2 \rangle$ to the dissipation, is absorbed into the model parameters, $P_b$ in Equation (3) is defined as the fraction of drops with turbulent kinetic energy greater than the drop surface energy. The structure function is used as an approximation of the turbulent kinetic energy imparted to the drop via eddy-drop interactions. The assumption here is that an oscillating deformed drop will break if the kinetic energy transmitted to the drop by the turbulence exceeds the drop surface energy. The authors postulate that the motion of drops and eddies is random, thus the fraction of the energetic interactions is expressed in terms of a two-dimensional Maxwell-Boltzmann distribution derived for an ideal gas.\textsuperscript{17}

Another widely used class of models,\textsuperscript{35-37} which follows a monotonic behavior with increasing drop size is the approach of Narsimhan et al,\textsuperscript{27} and its extensions by Alopaeus et al\textsuperscript{28} and Laakonen et al.\textsuperscript{38} The main assumption in this class is that drop fragmentation occurs if the relative velocity fluctuations between points local to the drop provide the minimum increase in the surface energy for breakage. The velocity fluctuations at the drop surface are thought to be due to the arrival of eddies of different scales, and it is postulated that the arrival of eddies at the surface of a drop is a stochastic process following a Poisson distribution. The Poisson parameter, which effectively denotes the frequency of the breakage event, is assumed to be independent of both the drop size and the flow field. Alopaeus et al’s,\textsuperscript{28} contribution to Narsimhan’s approach is two-fold; first, they include an energy dissipation rate dependence for the Poisson parameter; secondly, they account for the stabilizing viscous stresses according to Calabrese et al,\textsuperscript{39} as the original model is based on the assumption that viscous stresses are negligible. From this class of models, the model of Alopaeus et al\textsuperscript{28} (also used in the present study) is written as

$$b(\xi) = c_{1b}^{1/3} \text{erfc} \left( \frac{c_{2b}b}{\rho_c \sigma^{2/3}} + \frac{c_{3b}b}{\sqrt{\rho_d \sigma^{2/3}}} \right).$$

(4)

The breakage probability in Equation (4) is given by the complementary error function (erfc) term. A comparison with the breakage probability in Equation (3) shows an interesting similarity: the interfacial tension and viscous stress terms reported by both author groups are equal (for $\rho_c = \rho_d$, which is typical in emulsions). Both models are rooted to Hinze’s\textsuperscript{32} concept for the breakage criterion,\textsuperscript{17} comparing the turbulent kinetic energy at the drop length scale with the drop surface energy (the viscous term is obtained similarly).

The breakage frequencies of the Coulaloglou and Tavlarides\textsuperscript{21} and the Alopaeus et al\textsuperscript{28} models are summarized in Figure 1a. The model parameters and properties used in the figure correspond to a typical silicone–oil/water system and are given in Tables 1 and 2. Since $P_b$ is a monotonically increasing function of the drop size (Figure 1b) for both models, the non-monotonicity (or otherwise) of the models must stem from the different expressions for $t_b$ (Figure 1c).

A major challenge in drop breakage modeling is that the model parameters $c_{1b}$ - $c_{3b}$ are generally considered as system-specific, and are thus treated as fitting constants.\textsuperscript{17,20} Authors traditionally present their own parameter values aimed at reproducing experimental
results. For example, Maaß and Kraume reported that the parameter used in the Coulaloglou and Tavlarides model varies in the literature by approximately three orders of magnitude. The corresponding parameter appearing in the Alopaeus et al model exhibits an even larger variation of approximately five orders of magnitude. The wide variation of the parameter values highlights the large uncertainty introduced by the existing model formulations, which in turn affects the predictive capability of PBM.

In our previous work, we interpreted the breakage time in the model of Alopaeus et al via , which provides a unified approach to incorporate the underlying turbulence structure via . A generic power-law spectrum of the form

![Figure 1](image_url)

**TABLE 1** Physical and flow field properties used for the breakage frequency functions shown in Figure 1

| Property | Value |
|----------|-------|
| \( \mu_\text{a} \) [Pa s] | 0.01 |
| \( \rho_\text{a} \) [kg m\(^{-3}\)] | 1,000 |
| \( \mu_\text{c} \) [Pa s] | 0.001 |
| \( \rho_\text{c} \) [kg m\(^{-3}\)] | 1,000 |
| \( \sigma \) [N m\(^{-1}\)] | 0.01 |
| \( \varepsilon \) [m\(^2\) s\(^{-3}\)] | 1,000 |

**TABLE 2** Model parameters used for the breakage frequency functions shown in Figure 1

| Model | \( c_{1,b} \) [-] | \( c_{2,b} \) [-] | \( c_{3,b} \) [-] | \( c_{1,b} \) [m\(^{-2/3}\)] | \( c_{2,b} \) [-] | \( c_{3,b} \) [-] |
|-------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| Coulaloglou and Tavlarides Equation (3) | 0.00481 | 0.08 | 0.00785 | | | |
| Alopaeus et al Equation (4) | | | | 12 | 0.16 | 0.2 |
\[ E(\kappa) \propto \kappa^{-p}, \quad (5) \]

(where \( \kappa = 2x/r \)) corresponds to a second-order structure function of the form

\[ \langle \delta u^2 \rangle \propto r^q; \quad (6) \]

the exponents \( p \) and \( q \) are

\[
q = \begin{cases} 
  p - 1, & \text{if } p \leq 3 \\
  2, & \text{if } p > 3
\end{cases}
\quad (7)
\]

In this respect, the implied structure functions of the Coulaloglou and Tavlarides model\(^{21}\) and the Alopaeus et al model\(^{28}\) are \((p = 5/3, q = 2/3)\), and \((p > 3, q = 2)\), respectively.

The observation that the two models discussed here may be viewed as limiting responses to the underlying turbulence may be demonstrated using recent developments by Han et al\(^{42}\) and Solsvik et al\(^{30}\). In their respective treatments, a full spectrum is used based on homogeneous isotropic turbulence\(^{61}\)

\[ E(\kappa) = C_\varepsilon^2/3 \kappa^{-5/3} f_L(\kappa) f_\eta(\kappa \eta). \quad (8) \]

where

\[
f_L = \left( \frac{xL}{(xL)^2 + \alpha_1} \right)^{11/3} \quad (9)
\]

and

\[
f_\eta = \left( \frac{xL}{(xL)^2 + \alpha_1} \right)^{11/3} \quad (9)
\]

FIGURE 2  (a) Energy spectrum from Equation (8); (b) structure function from Equation (11); (c) breakage times and; (d) breakage frequency functions from Equations (3), (4), and (12) for the parameters included in Tables 1 and 2 and \( Re_L = 10^5 \). The straight lines in panels (a) and (b) represent the assumed slopes of the C&T (inertial subrange) and Alopaeus et al models from Equations (5)–(7) [Color figure can be viewed at wileyonlinelibrary.com]
\[ \eta = \exp\left(-\beta_0 \kappa \eta^4 + c_\eta^4 \right)^{1/4} - c_\eta \]  

4 + c_\eta^4 / \eta^{18/29} \] 

43. If we calculate the second-order structure function for this spectrum 44 

\[ \frac{\delta u^2}{\zeta} = \frac{3}{2} \exp\left(-c_2 \beta_2 \frac{\delta u^2}{\rho \xi^2 (\delta u^2)^2} - c_3 \beta_3 \frac{1}{\rho \xi^2 (\delta u^2)^2} \right). \]  

As shown in Figure 2d, the main effect of accounting for the full turbulence spectrum is that the breakage frequency in both the small and the large scales is smaller than that in the inertial subrange, due to the larger breakage times in these regions. The decrease in the small scales is observed for \( \xi/\eta \lesssim 15 \), and is driven by the decrease of the breakage time slope from 2/3 to 0 shown in Figure 2c. 

The extension of drop breakage modeling to the full spectrum presents a significant development which potentially reduces experimental costs required to refit the parameters in prior breakage frequency models. 20,30 However, the assumptions of homogeneity and isotropy used are still questionable. Solsvik et al. 30,46 argued that the effect of anisotropy is negligible compared to the effect of finite Reynolds number on breakage frequency; however, this view was not explored in further detail. As it will be discussed in the last section where we compare breakage frequencies based on isotropic and anisotropic spectra, the relative influence of anisotropy on breakage frequency increases with decreasing Reynolds number. 

3 | BREAKAGE FREQUENCY USING THE FULL SPECTRUM BASED ON HOMOGENEOUS ISOTROPIC TURBULENCE

To obtain the breakage frequency for the full spectrum, one may use the Couloaglou and Tavlarides approach by replacing \( \rho \zeta^2/3 \) in the original model (Equation (3)) with \( \langle \delta u^2 \rangle \) from Equation (11). 45 The breakage frequency is then written as

\[ b(\xi) = c_{1b} \frac{\sqrt{\langle \delta u^2 \rangle}}{\xi} \exp\left(-\frac{c_{2b} \sigma_1}{\rho_1 \xi^2 (\langle \delta u^2 \rangle)^2} - \frac{c_{3b} \mu_1}{\rho_1 \xi^2 \sqrt{\langle \delta u^2 \rangle}} \right). \]  

FIGURE 3  (a) Perturbed \( (E_{11,ani}) \) and unperturbed \( (E_{11}) \) energy spectra from Equations (13) and (23), respectively, for \( Re_\theta = 10^5 \) and the parameters included in Table 1. For \( E_{11,ani} \): \( A = 16, \kappa_1 = 2\pi/L_11, L_11 = 0.43L, \) and \( \sigma_1 = \kappa_1/10, E_{11,ani} \) integrates to the same \( k \) as \( E_{11} \). The vertical line denotes the low wave number limit of the inertial subrange. (b) Structure functions from Equation (14) corresponding to \( E_{11,ani} \) and \( E_{11} \). The vertical line denotes the upper length scale limit of the inertial subrange [Color figure can be viewed at wileyonlinelibrary.com]
in the corresponding second-order structure functions is thus masked by vortex shedding and the resulting structure functions exhibit slopes between 2/3 and 2 in the inertial subrange. In a separate study of a rotor-stator mixer, PIV-measured 1-D energy spectra at the stator slot jet reveal deviations from model spectra based on HIT. The deviations manifest themselves as steeper than \(-5/3\) slope in the low wave number end of the inertial subrange and a narrow range of wave numbers with \(-5/3\) slope. A previous PIV study of the same rotor-stator configuration identified periodic vortex shedding inside the stator slot due to the rotor blade passage. The authors in Reference concluded that the deviations between the measured spectra and the model spectra are consistent with finite Reynolds number effects (with \(Re_L \approx 1000 - 3000\)). In the high-pressure homogenizer study, \(Re_L\) was also moderate ranging from \(-10^3 - 10^4\).

The amplification of energy in these studies is restricted to the energy-containing range and the low wave number end of the inertial subrange; its effect, however, on the second-order structure function extends to a much broader range of scales, as demonstrated by Portela et al. By assuming a 1-D model energy spectrum of the form

\[
E_{11,\text{an}}(\kappa_1) = \frac{1 + A \exp \left( - \frac{\kappa_1^2}{2\sigma_1^2} \right)}{\kappa_1^2} E_{11}(\kappa_1),
\]

(13)

(where \(\sigma_1\) represents the spread of the spectral peak, \(A\) is an amplification factor and \(E_{11}\) is the 1-D model spectrum), the second-order structure function was calculated as

\[
\langle \delta u^2 \rangle = 2u^2 \left[ 1 - \mathcal{F}^{-1} (E_{11,\text{an}}) \right]
\]

(14)

where \(\mathcal{F}^{-1}\) denotes an inverse Fourier transform. Figure 3a shows a perturbation restricted to the energy-containing range. The low wave number limit of the inertial subrange is indicated by the vertical line.

**FIGURE 4** (a) 1-D energy spectra from LES and the model Equation (22) for local values of \(Re_L, \epsilon, \frac{C_2}{C_1}\) obtained from LES, \(\kappa_1 = 2\pi/L_1\) and \(\sigma_1 = \kappa_1/5\); (b) largest stress anisotropy component from LES; and (c) turbulence Reynolds number from LES. Results shown here correspond to different streamwise locations of a Sonolator jet half-velocity width [Color figure can be viewed at wileyonlinelibrary.com]
The main effect on the structure function, shown in Figure 3b, is an increased slope over a much broader range of length scales. The figures clearly demonstrate how a perturbation outside the inertial subrange can influence the dynamics within the subrange (and hence impinge on the validity of older breakage models).

5 ENERGY SPECTRUM–STRUCTURE FUNCTION MODEL BASED ON TURBULENCE ANISOTROPY

5.1 Model derivation

In this section we develop the ideas reported in Portela et al.51 to model the second-order structure function by perturbing the underlying energy spectrum via turbulence anisotropy. Our model is designed to be used with Reynolds-stress transport models which account for the individual Reynolds stresses (and hence the stress anisotropy) directly.

The departure from isotropy can be expressed in terms of the deviatoric part of the Reynolds stress tensor,21:

\[ \overline{u'u'} = \frac{2}{3} \kappa \delta + \left( \overline{u'u'} - \frac{2}{3} \kappa \delta \right). \] (15)

Assuming that the anisotropy is confined to the large scales and considering only the largest principal stress \( \overline{u''_1} \in [2k, 2k] \),52 then we model the 1-D energy spectrum according to Equation (13). \( \kappa_1 \) and \( \sigma_1 \) are suitably chosen such that the perturbation is confined to the energy-containing range. We assume that the contribution to \( \overline{u''_1} \) arising from anisotropy is equal to the amount of energy added to the spectrum via the perturbation. \( \overline{u''_1} \) is thus written as

\[ \overline{u''_1} = \int_{0}^{\infty} E_{11}(\kappa_1) d\kappa_1 + A \int_{0}^{\infty} \exp \left( -\frac{(\kappa_1 - \overline{\kappa})^2}{2\sigma_1^2} \right) E_{11}(\kappa_1) d\kappa_1. \] (16)

Since

\[ \int_{0}^{\infty} E_{11}(\kappa_1) d\kappa_1 = \frac{2}{3} k \] (17)

it follows that

\[ A = \frac{1}{f_A} \left( \frac{\overline{u''_1}}{k} - \frac{2}{3} \right). \] (18)

where

\[ f_A = k^{-1} \int_{0}^{\infty} \exp \left( -\frac{(\kappa_1 - \overline{\kappa})^2}{2\sigma_1^2} \right) E_{11}(\kappa_1) d\kappa_1. \] (19)

\( f_A \) depends on \( Re, \kappa_1, \) and \( \sigma_1 \) (but not on stress anisotropy). Equation (18) states that the perturbation parameter \( A \) is proportional to the departure from turbulence isotropy. To ensure the perturbed spectrum integrates to the same total turbulent kinetic energy as the unperturbed spectrum, Equation (16) is rescaled as follows

\[ \int_{0}^{\infty} \frac{1}{A_1} \left[ 1 + \exp \left( -\frac{(\kappa_1 - \overline{\kappa})^2}{2\sigma_1^2} \right) \right] E_{11}(\kappa_1) d\kappa_1 = \frac{2}{3} k. \] (20)

In the Supporting Information it is shown that

\[ A_1 = \frac{3\overline{u''_1}}{2k}. \] (21)

and hence, the proposed normalized perturbation to the 1-D energy spectrum is finally written as

\[ E_{11,ani}(\kappa_1) = \frac{2k}{3\overline{u''_1}} \left[ 1 + \frac{1}{f_A} \left( \frac{\overline{u''_1}}{k} - \frac{2}{3} \right) \exp \left( -\frac{(\kappa_1 - \overline{\kappa})^2}{2\sigma_1^2} \right) \right] \] (22)

The energy associated with the perturbation is assumed to be normally distributed around a mean wave number \( \overline{\kappa} = \frac{4L}{\pi} \) with a SD \( \sigma_1 \). \( L_{11} \) is the longitudinal integral length scale which depends on \( Re \) and decreases asymptotically to 0.43L with increasing \( Re \). \( \sigma_1 \) is chosen such that the perturbation is contained in the energy-containing range \( [\overline{\kappa}, \frac{12\pi}{L}] \).51 \( f_A \) is calculated from Equation (19) and the 1-D model spectrum from51

\[ E_{11}(\kappa_1) = \int_{E_1}^{\infty} \frac{E(\kappa)}{\kappa} \left[ 1 - \left( \frac{\kappa_1}{\kappa} \right)^2 \right] d\kappa. \] (23)

For given \( k, \kappa, \overline{u''_1} \) and the above assumptions about \( \kappa_1 \) and \( \sigma_1 \). Equation (22) returns a spectrum shape which mimics the 1-D spectra observed in our LES of a high-pressure homogenizer.47 This is achieved by redistributing energy from across the full spectrum to the wave numbers local to the vortex shedding frequency. The model may thus be considered as a specific realization of turbulence anisotropy pertinent to fluid flows dominated by vortex shedding. It is noted that the model is not limited to the configuration studied in our LES: any modeled anisotropy from any known source can in principle be accounted. The second-order structure function is obtained via the Fourier transform of the perturbed spectrum by41

\[ \langle \delta u^2 \rangle = 2 \int_{0}^{\infty} E_{11,ani}(\kappa_1) \left[ 1 - \cos(\kappa_1 r) \right] d\kappa_1. \] (24)

5.2 Model assumptions

To describe the evolution of energy spectra and second-order structure functions, a model able to capture spectral dynamics is
For example, the equation derived by Monin and Yahlom is exact in describing the evolution of an energy spectrum in homogeneous and anisotropic turbulence with imposed mean velocity gradients. Then, the structure function related to the energy spectrum by a Fourier transform can be used to evaluate breakage frequency. A different approach is to directly account for the evolution of structure functions. The Karman–Howarth–Monin–Hill (KHMH) equation is a general model without the need for making any assumptions about inhomogeneity and anisotropy. There are several works where the various terms of the KHMH equation are assessed (by post-processing either DNS or experimental data) for their respective contributions to the evolution of the structure functions and to the turbulence cascade.

To our knowledge, a model mimicking the essential features of either the Monin-and-Yahlom or the KHMH equation is not available. We have thus restricted ourselves to a specific shape of the spectrum and a simple algebraic expression to link anisotropy to the perturbation in the spectrum which is tractable for use in CFD-PBM simulations.

5.3 Comparison of model spectra with LES spectra

Figure 4 depicts 1-D energy spectra extracted from our LES in a Sonolator high-pressure homogenizer and model spectra from Equation (22). From Figure 4a it may be seen that the model spectra describe qualitatively the amplitude and the mean wave number of the energy perturbation at the various locations. The location of the probes where the spectra are evaluated correspond to the half-velocity widths of the shear layer at the near field of the Sonolator jet. Z represents the streamwise distance from the jet exit, normalized with the nozzle hydraulic diameter. The dissipation rate of turbulent kinetic energy peaks at $Z/D_h = 0.5 - 2$, therefore the bulk of drop breakage is expected to occur in that region. The largest principal Reynolds stress in these locations is $u_{\tau}^2$. The stress-anisotropy component shown in Figure 4b, reduces rapidly with increasing streamwise distance; in the region of peak dissipation rate the magnitude of $a_{33}$ remains considerable. $Re_c$ at the same locations (shown in Figure 4c) is $O(10^7 - 10^8)$.

5.4 Comparison of structure functions based on 1-D and 3-D energy spectra

Before implementing the proposed model (Equations (22)-(24)) into a breakage frequency model it is instructive to examine its behavior in the limit of isotropic turbulence. In the isotropic limit, $u_{\tau}^2 = 2/3$, and Equation (22) simplifies to Equation (23). We can then compare this with the 3-D energy spectrum—structure function model in HIT (Equation (8) and (11)). Figure 5a shows a 3-D spectrum from Equation (8) and a 1-D spectrum from Equation (22) for $u_{\tau}^2 = 2/3$. The corresponding structure functions, obtained from Equations (11) and (24), respectively, are shown in Figure 5b; the structure functions coincide for the entire dissipation range and inertial subrange. There is a minor difference in the energy-containing range shown in the inset of Figure 5b which, however, is relatively less important as the DSDs typically lie within the dissipation range and the inertial subrange. We thus conclude that the isotropic limit of our 1-D energy spectrum—structure function approach using Equations (23) and (24) is equivalent to the HIT 3-D energy spectrum—structure function approach.
Comparison between isotropic and anisotropic models

We now examine the effect of turbulence anisotropy on breakage time and frequency by implementing the 1-D energy spectrum—structure function in a breakage frequency model. The breakage frequency is calculated in the same manner as with other full spectrum approaches (Equation (12)). Figure 6 shows energy spectra, structure functions, breakage times and frequencies for different values of \( u'^2_1 / \kappa \) and \( \text{Re}_L = 2,000 \), using the parameters included in Tables 1 and 2. According to Pope, the length scales of the inertial subrange extend from \( 60 \eta \) to \( \frac{L}{2} \). These values apply to very-large \( \text{Re}_L \); since Pope’s limits overlap for small \( \text{Re}_L \), \( \eta \frac{L}{2} \) has been suggested for \( \text{Re}_L \approx 8630 \). For \( \text{Re}_L = 2000 \), the inertial subrange in Figure 6 is identified as \( r_1 \in [15 \eta, 80 \eta] \). The main effect of increasing \( \frac{u'^2_1}{\kappa} \) on the structure function is to increase the slope toward a value of 2 in the range \( \sim [15 \eta, 80 \eta] \). In this range, the breakage time exhibits a decreasing slope with increasing anisotropy from the isotropic value of 2/3 to \( \sim 0 \). The isotropic scaling in the inertial subrange moves toward the implied breakage time term in the model of Alopaeus et al.\(^{28}\) In the same range, the breakage frequency increases with increasing anisotropy, driven by the reduced breakage time. Thus, for finite Reynolds number flows, the effect of anisotropy on breakage frequency is implicitly embodied in the model of Alopaeus et al.\(^{28}\)

In the small length scales of the inertial subrange, the breakage time of the full spectrum isotropic approach (Equations (11) and (12)) and that of the Alopaeus et al model\(^{28}\) may be viewed as two extremes of the length scale exponent (\( 2 \) and 0, respectively) since from Equations (2) and (6)

\[
\frac{t_b}{\xi} \sim \left( \frac{\xi}{\sqrt{\langle \delta u^2 \rangle}} \right) \sim \frac{\xi}{\sqrt{\psi}} \sim \xi^{2-\epsilon/2}
\]

(26)
where \( q \leq 2 \) (Equation (7)) is the slope of the structure function. Our model provides a physically plausible mechanism which predicts these extremes as well as intermediate values of the exponent, subject to the local \( R_e_L \) and anisotropy. It is worth drawing a comparison here with the work of Håkansson et al\(^{59,60} \) who obtained experimentally turbulent energy spectra in the outlet chamber of a high-pressure homogenizer and attempted to fit a characteristic breakage velocity. To account for the increased slope of the spectrum observed in their measurements, they proposed a form \( h \delta u^2 / \xi^{1.3} \) (i.e., \( t_b / \xi^{0.35} \) from Equation (26)), which suggests an intermediate state between strong turbulence anisotropy and homogeneous isotropic turbulence.

Figure 7 explores the dependence of structure function, breakage time and breakage frequency on \( R_e_L \). The main point to note here is that, for large \( R_e_L \), the shape of the breakage time curve (hence the breakage frequency) in the small scales of the inertial subrange remains unaffected by anisotropy.

**5.6 Comparison between anisotropic and intermittency models**

In the previous section it was shown that the introduction of anisotropy via a perturbation to the energy spectrum alters the slope of the structure function in the inertial subrange, thereby affecting the shape of the breakage frequency curve. A different phenomenon which also has an impact on that slope is intermittency. In this section, we compare the proposed anisotropic model with an intermittency model in order to assess their relative importance.

Intermittency of turbulence refers to spatial fluctuations of quantities based on velocity differences or velocity gradients, for example vorticity or energy dissipation rate. Based on the scale of the fluctuations, intermittency is distinguished as external or internal.\(^{44} \) External intermittency is related to the large scales and is not considered to be universal. Internal intermittency is related to the small scales and is considered to affect the universality of small-
scale statistics. The spotty structure of velocity-gradient based quantities can be described by considering their respective PDFs. While velocity measured at a single point follows a nearly gaussian distribution, that is, it behaves as a random variable, the velocity gradient PDFs are strictly non-gaussian and their flatness factor is larger than the gaussian value of 3. This non-normal behavior is essential to the dynamics in turbulence, and the underlying physical mechanism is vortex stretching.41

On the grounds of intermittency, Landau44 challenged the universality of small-scale statistics advanced by Kolmogorov’s theory, motivating the development of the revised similarity hypotheses.61,62 The influence of internal intermittency on the structure function can be accounted for through a modification of the power law exponent, that is, \( \langle \Delta u^p \rangle \propto r^{n+2} \). Among the various intermittency models, the log-normal model by Kolmogorov and Obukhov61,62 may be used to obtain the revised exponent as

\[
\zeta_n = \frac{n}{3} + \frac{\mu}{18} \ln(n+2)
\]  

where \( \zeta_n \) is the revised exponent in \( \langle \Delta u^p \rangle \propto r^{\zeta_n} \) and \( \mu = 0.25 \) is a model parameter fitted with experimental data.61 The correction is considerable for higher-order structure functions. The revised exponent of the second-order structure function, relevant to droplet breakage modeling, is \( \sim 4\% \) larger than the value predicted by the original theory. This is considered a small change taking account of the other uncertainties involved in droplet breakage.63,64

Meyers and Meneval65 introduced the effects of intermittency by modifying Pope’s 3-D model spectrum.41 Their model is written as

\[
E(\kappa) = C_k \kappa^{-5/3} (xL)^{-\beta} f\kappa(xL) f_\kappa(x\eta),
\]  

where
\[ f_L = \left( \frac{\kappa L}{(\kappa L)^{1/2} + \alpha_5} \right)^{11/3 + \beta} \]  

(29)

and

\[ f_n = \exp(-\alpha_1 \kappa \eta) B(\kappa \eta) \]

(30)

are reshape functions for the energy containing range and the dissipation range, respectively, and the factor

\[ B(\kappa \eta) = \left[ 1 + \frac{\alpha_2 (\kappa \eta / \alpha_4)^{\alpha_2}}{(1 + \kappa \eta / \alpha_4)^{\alpha_2}} \right] \]

(31)

is a bottleneck correction used to shape the spectral bump on the onset of the dissipation range; \( \beta = \mu / 9 \) is the intermittency correction for the inertial subrange and \( \mu = 0.25 \), defined in Equation (27); \( \alpha_1 \) and \( \alpha_2 \) are model parameters, fitted with experimental data, and we use their respective values reported by Solsvik and Jakobsen.\(^64\)

An intermittency-corrected 1-D spectrum is obtained using Equation (23) by numerically integrating the 3-D spectrum, Equation (28), similar to the approach of Meyers and Menevau.\(^65\) The result is shown in Figure 8a along with isotropic and anisotropic 1-D spectra. Comparing to the baseline isotropic spectrum, the intermittency model redistributes energy to the high wavenumbers of the inertial subrange, to account for the bottleneck effect, while the anisotropy model redistributes energy to the energy containing range. The amount of redistributed energy with the anisotropy model is much larger than that with the intermittency model, even though we have used a moderate value for the stress anisotropy, \( \frac{\tau_{ij}}{\tau} = 1 \). The corresponding structure function, breakage time and breakage frequency based on the intermittency-corrected 1-D spectrum are calculated using Equations (24), (2), and (12), respectively, and illustrated in Figure 8b–d. The main point to note here is that the effect of intermittency to the shape of the breakage frequency curve is significantly smaller than the effect of anisotropy.

6 | CONCLUSION AND FUTURE WORK

The majority of turbulent drop breakage in emulsification devices is expected to occur in locations where turbulence production and dissipation peak. These regions are characterized by turbulence anisotropy and deviations of the energy distribution from model spectra assuming homogeneous isotropic turbulence.

We have presented a novel approach aiming to capture the effect of a specific realization of turbulence anisotropy on the second-order structure function used in drop breakage modeling. The specific realization is related to spectral peaks and slopes in the inertial subrange steeper than Kolmogorov’s \(-5/3\) scaling, which have been observed in industrially relevant geometries. Our approach incorporates the recent extension\(^30\) of drop breakage modeling to the full spectrum of isotropic turbulence, as a limiting case of zero anisotropy. By using the new structure function in the model of Coulaloglou and Tavlarides,\(^21\) we have shown that the main effect of anisotropy is to alter the non-monotonic behavior of an HIT-based breakage frequency toward a monotonic one; this effect is more pronounced for smaller turbulence Reynolds numbers. Apart from drop breakage processes, the proposed model may also be used in rate functions of other particulate processes related to the turbulent stresses, such as drop coalescence.

In future work, this model is to be implemented in a CFD solver and used in CFD-PBM simulations of turbulent emulsification in high-pressure homogenizers. Instead of two-equation RANS models, typically used in such studies, a Reynolds-stress transport model is employed to account for turbulence anisotropy, which is then introduced in drop breakage modeling through Equations (22) and (24).

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AUTHOR CONTRIBUTIONS

Ioannis Bagkeris: Conceptualization; methodology; writing-original draft; writing-review and editing. Vipin Michael: Conceptualization; methodology; supervision; writing-original draft; writing-review and editing. Robert Prosser: Conceptualization; funding acquisition; methodology; project administration; resources; supervision; writing-original draft; writing-review and editing. Adam Kowalski: Funding acquisition; project administration; resources; supervision.

DATA AVAILABILITY STATEMENT

Data sharing not applicable to this article as no datasets were generated or analysed during the current study.

NOTATION

C&T model of Coulaloglou and Tavlarides
CFD computational fluid dynamics
HIT homogeneous isotropic turbulence
LES large-eddy simulation
PBE population balance equation
PBM population balance modeling
PDF probability density function
QMOM quadrature methods of moments
RANS Reynolds-averaged Navier–Stokes

GREEK SYMBOLS

\( \eta \) Kolmogorov length scale, m
\( \kappa \) magnitude of wave number vector, m\(^{-1}\)
\( \kappa_i \) wave number component in the \( i \)th direction, m\(^{-1}\)
\( \mu \) dynamic viscosity, Pa s
\( \varepsilon_{ij} \) energy perturbation mean wave number, m\(^{-1}\)
\( \varepsilon_{ij} \) energy perturbation standard deviation
\( \rho \) density, kg m\(^{-3}\)
\( \sigma \) interfacial tension, N m\(^{-1}\)
ε  
  turbulent kinetic energy dissipation rate, m² s⁻³

ξ  
  drop diameter, m

ROMAN SYMBOLS

(\bar{\alpha}u_i^2)  
  second-order structure function, m² s⁻²

b  
  drop breakage frequency, s⁻¹

c_{1,b} - c_{3,b}  
  breakage model parameters

Dh  
  hydraulic diameter, m

E(k)  
  3-D energy spectrum in HIT, m³ s⁻²

E_{i,j}(k)  
  1-D energy spectrum of u_i in the jth direction, m³ s⁻²

k  
  turbulent kinetic energy, m² s⁻²

L  
  integral length scale, m

L_{11}  
  longitudinal integral length scale, m

P_b  
  drop breakage probability

ρ  
  scalar distance between two points, m

ReL  
  turbulence Reynolds number

t_b  
  drop breakage time, s

u_i  
  fluctuating velocity component in the i-th direction, m s⁻¹

SUBSCRIPTS

ani  
  anisotropic

C  
  continuous phase

d  
  dispersed phase

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