DROP BREAKUP AND INTERMITTENT TURBULENCE

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Key Words: Drop Size, Energy Dissipation, Inertial Sub-range, Intermittency, Multifractal, Scale Effect

Recent studies on the fine structure of turbulent flow are applied to drop breakup in the inertial sub-range. A multifractal method describes intermittency and the distribution of velocity fluctuations etc. For a given drop size and a given time-averaged energy dissipation rate, a wide range of stresses acts to cause breakup. These stresses and their relative frequencies are calculated. The most likely exponent on the Weber Number is close to –0.6. Smaller values (possibly as low as –0.93) arise from rare, but violent intermittent turbulence. Such low exponents are likely after long agitation times and for small tanks.

Introduction

A bubble or drop, suspended in a continuous phase, will break up if the stress exerted by motion in the continuous phase exceeds the stabilizing forces due to the surface tension and the drop viscosity. For drops of low viscosity, whose maximum diameter falls within the inertial sub-range of the continuous phase turbulence and whose volume fraction is so low that coalescence does not occur, the following proportionality can be derived theoretically[9)]

$$d^0_{\max} \sim \sigma^{0.6}\,\varepsilon^{-0.4}\,\rho^{-0.6} \tag{1}$$

where σ is the interfacial tension, ε the rate of dissipation of turbulent kinetic energy, ρ the density of the continuous phase and d^0_{max} the diameter of the largest stable drop, predicted by ignoring turbulence intermittency. Expressed in dimensionless form this becomes

$$\frac{d^0_{\max}}{D} \sim We^{-0.6} \tag{2}$$

where D is a characteristic linear dimension and *We* is the Weber Number, which will subsequently be treated in more detail.

Many measurements of drop sizes e.g. see Refs.[5, 17, 18)] agree quite well with Eq. (2). It has also been found that Eq. (2) may be extended to include the influence of the drop viscosity[5, 19, 18)] and that the Sauter mean diameter is usually proportional to d_{max}, so that d_{32} is also correlated by Eq. (2).

Despite the usefulness of Eq. (2) in correlating drop size measurements, some recent studies have given smaller exponents than –0.6. For example results from three tank sizes[11)] followed Eq. (3)

$$d_{\max} \sim D^{-1.5}\,N^{-1.6} \tag{3}$$

where N is the rotational speed of a Rushton turbine. The exponents on N and D would be –1.2 and –0.8 in order to be consistent with the exponent –0.6 on *We*. The results in Eq. (3) imply exponents between –0.8 and –0.83 however. In another study[4)] the exponents on *We* for different impellers were close to –0.7. Although the Reynolds Numbers were rather low, well-developed turbulence should have existed near the impellers. In some experiments the drop sizes neared the Kolmogorov scale, although it seems this has no effect on the exponent[13)]. {Well within the viscous sub-range the exponent should become –1}. In a third case[12)] the exponent was –0.93 for low-viscosity drops. A mechanism applying when $d_{max} \approx L$, where L is the integral scale or size of the energy-containing eddies, has been put forward[11)], which results in an exponent –1 on *We*.

It is the purpose of this contribution to show that the classical, inertial sub-range theory[9)] can be modified by including the effect of intermittency to yield exponents smaller than –0.6 in certain circumstances and equal to –0.6 in others. This unified theory can not yet be regarded as validated by experiments and suggestions to this end are therefore also included.

1. Intermittency

Intermittency refers to random variations in turbulence properties at a fixed point in a turbulent flow. Here fluctuations in the fine-scale properties—in particular the energy dissipation rate, ε—are central. Whereas intermittency is an established experimental phenomenon, its modelling still gives rise to difficulties. Kolmogorov[10)] suggested that log ε is normally distributed, but this is inadequate for the high –ε tail of the distribution. The β-model[7)] attributes the whole dissipation at a particular instant to active parts of the whole space, whilst the remainder is non-turbulent. Thus ε has either a constant value or is zero. This has didactic value, is however over-simplified. Application[1)] to rapid drop breakup yields exponents on We smaller than –0.6 and it is there-

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fore worth applying a more realistic intermittency model to this problem. Whereas the β-model rests on a single (fractal) dimension for the fine structure, it is better to work with a distribution of such dimensions, termed a multifractal model, whose parameters have been carefully measured. An outline of the main results, relevant to drop breakup, will now be given.

When the length and velocity vectors, $\bar{r}$ and $\bar{v}$, the time t and the pressure P arising in the Navier-Stokes equation are transformed according to

$$\bar{r}' = \lambda \bar{r} \tag{4}$$

$$\bar{v}' = \lambda^{\alpha/3} \bar{v} \tag{5}$$

$$t' = \lambda^{1-\alpha/3} t \tag{6}$$

$$\left(\frac{P}{\rho}\right)' = \lambda^{2\alpha/3}\left(\frac{P}{\rho}\right) \tag{7}$$

then the Navier-Stokes equation is invariant[6, 16]. (The inertial sub-range must include $\bar{r}$ and $\bar{r}'$).The exponent a is arbitrary and its distribution of values has been determined experimentally[15].

The velocity difference v_r between points separated by a distance r follows from Eq. (5)

$$\frac{v_r}{v_L} = \left(\frac{r}{L}\right)^{\alpha/3} \tag{8}$$

In this and all following equations, $\alpha = 1$ corresponds to the classical, non-intermittent relationships for the inertial sub-range. v_L is the velocity fluctuation of the energy-containing eddies. Now

$$\langle \varepsilon \rangle = v_L^3 / L \tag{9}$$

where $<\varepsilon>$ is the time-averaged energy dissipation rate. Denoting over a domain of size r the energy dissipation rate by ε_r, it may be expected that

$$\varepsilon_r = v_r^3 / r \tag{10}$$

Substituting from Eq. (8) and (9), Eq. (10) becomes

$$\varepsilon_r = \langle \varepsilon \rangle \left(\frac{r}{L}\right)^{\alpha - 1} \tag{11}$$

Again $\alpha = 1$ signifies no intermittency and a uniform energy dissipation rate.

The derivation of the maximum stable drop size, d_{max}, now parallels exactly the non-intermittent case[5, 9, 17]. The turbulent stress acting upon a drop of size r, where r is in the inertial sub-range, is

$$\tau(r) \sim \rho v_r^2 \sim \rho\,[\langle \varepsilon \rangle r]^{2/3} \left(\frac{r}{L}\right)^{2(\alpha-1)/3} \tag{12}$$

after substituting from Eq. (8) and (9). This stress will be just equal to the stabilising stress σ/d_{max} when $r = d_{max}$, so that from (12)

$$d_{\max} \sim \left(\frac{\sigma}{\rho}\right)^{1/\{1+2\alpha/3\}} \varepsilon^{-2/3\{1+2\alpha/3\}} L^{-2\{(1-\alpha)/3\}/\{1+2\alpha/3\}} \tag{13}$$

Comparing with Eq. (1), Eq. (13) reduces to the well-known result when $\alpha = 1$, but otherwise there will be an effect of scale, since L is proportional to linear size e.g. for a Rushton turbine $L \approx 0.1\ D$, where D is the turbine diameter. For a stirred tank We is defined by

$$We = N^2 D^3 \rho / \sigma \tag{14}$$

and Eq. (13) may be re-arranged to give

$$\frac{d_{\max}}{D} \sim We^{-0.6a} \tag{15}$$

$$a = [1 - 0.4(1-\alpha)]^{-1} \tag{16}$$

α is a distributed quantity. Referring to Eq. (8), this means that for a given separation (or drop size) r, the velocity difference or increment, v_r, is also distributed, whereas in the classical case of no intermittency, v_r takes just one value. In Eq. (12) $\tau(r)$ also possesses a distribution, and so does d_{max}. The probability density function for α, $P(\alpha)$, is in general given by[16]

$$P(\alpha) \sim \left(\frac{r}{L}\right)^{d_s - f(\alpha)} \tag{17}$$

where d_s is a space dimension and $f(\alpha)$ is the multifractal spectrum, whose measurements for $d_s = 1$ in the range $0.51 < \alpha < 1.78$ have been correlated by[15, 16]

$$f(\alpha) = D_0 - \frac{(\alpha - \alpha_0)^2}{4(\alpha_0 - D_0)} \tag{18}$$

In Eq. (18) $D_0 = 1$ and $\alpha_0 = 1.117$ giving

$$P(\alpha) \sim \left(\frac{r}{L}\right)^{2.137(\alpha - 1.117)^2} \tag{19}$$

The low - α tail of the $P(\alpha)$ distrbution has been approximated by a square-root-exponential relationship[15]. Since $(r/L) < 1$, $P(\alpha)$ has a maximum when $\alpha = 1.117$, whereas when $\alpha << 1$, $P(\alpha)$ is much smaller. From Eq. (11), such low values of α correspond to peaks in ε, where $\varepsilon >> <\varepsilon>$, but such violent events are rare *i.e.* $P(\alpha)$ is small. Because the width of the inertial sub-range increases with rising Reynolds Number, e.g. when scaling up at constant power per unit volume ($<\varepsilon>$ = constant), the bursts of high ε become more violent, but also less frequent. This corresponds to the experimentally well known "spotty" distribution of the energy dissipation rate.

2. Application to Drop Breakup Experiments

Depending upon the value of α, Eq. (12) expresses the range of stresses acting upon a drop of diameter r in the inertial sub-range. Eq. (17) gives the probability of a particular α value (all possible α values occur in the turbulence at all times). The weighted stress in a given turbulent flow when α is close to 1 is therefore

$$\rho\,[v_r(r, \alpha)]^2 P(\alpha) \sim \left(\frac{r}{L}\right)^{(2\alpha/3) + 2.137(\alpha - 1.117)^2} \tag{20}$$

The most likely weighted stress corresponds to the smallest exponent in Eq. (20) since $(r/L) < 1$, which gives $\alpha = 0.961$. From Eq. (15) and (16), maximum as well as Sauter mean diameters should often be correlated by

$$\frac{d_{\max}(\text{or } d_{32})}{D} \sim We^{-0.61} \tag{21}$$

The result means that, when taking turbulent intermittency into consideration, the most likely result is nearly equal to that neglecting intermittency (–0.6) and has frequently been obtained by experimentalists.

The smallest value of α consistent with known experimental results[15] is 0.12, which predicts

$$\frac{d_{\max}\,(\text{or } d_{32})}{D} \sim We^{-0.93} \tag{22}$$

which has also been obtained from drop breakup experiments[12, 13]. This value of α corresponds to violent, but relatively rare bursts of energy dissipation. The ratio of maximum drop size to the integral scale of turbulence in these experiments[11, 13] was on the order of 0.001 ÷ 0.01. From Eq. (17) the ratios of the probabilities of the least likely and most effective α values (0.12 and 0.961) gives

$$\frac{P(\alpha=0.12)}{P(\alpha=0.961)} = 2\times10^{-5}\left\{\frac{d_{\max}}{L}=0.01\right\}$$
$$\text{and } 3\times10^{-9}\left\{\frac{d_{\max}}{L}=0.001\right\} \tag{23}$$

Because violent turbulence is rare —see Eq. (23)— exponents as small as –0.93 are likely to be influential only after long periods of time and for small tanks (d_{max} / L not too small, remembering that L~D). Short contact times on the order of seconds characterise turbulent flows in pipes[8] and static mixers[2] where drop sizes are consistent with an exponent of –0.6 on We. In stirred tanks, drop breakup occurs near the Rushton turbine[11], e.g. in the trailing vortices, where $<\varepsilon>$ is higher than elsewhere. All drops to be broken need to pass through this region, whose volume fraction (active volume / tank volume) is denoted by X. The mean circulation times for Fig. 3 and Fig. 4 of reference 11) were at most around 8 s and 2 s respectively. Circulation times of individual fluid elements have a log normal distribution[3], and some fluid might have needed an order of magnitude more time than the mean circulation times. Significant reductions in drop size continued to take place between 1/2 h and 5 h in Fig. 3 and between 1 h and 5 h in Fig. 4. These long times can hardly be explained by the circulation times, but probably reflect the slow shift in drop sizes from those corresponding to the most effective turbulence to those determined by more violent, but less frequent events. Seen in the light of intermittent turbulence, a gradual decrease in drop size (following the initial formation of a dispersion) together with a decrease in the exponent on We from –0.61 to –0.93 are to be expected. For scale-up at constant power per unit volume in order to attain given values of d_{max} and d_{32}, it is less probable that the asymptotic drop sizes and exponents will be attained in large tanks —compare Eq. (23)— within the duration of an experiment.

Eq. (13) was derived by balancing the disruptive and cohesive stresses, without any reference to time: it is a quasi-steady state balance. The time evolution of the drop size distribution can be calculated by solving the population balance, incorporating appropriate breakage relationships[14]. The influence of intermittency can be deduced semi-quantitatively as follows.

The product of number of passes of the dispersion through the active zone during a given period and the residence time in this zone is independent of stirrer speed and scale. It follows that the frequency of such passes multiplied by their duration equals X. Eddies of size r have a mean frequency (inverse of lifetime or turnover time) of $<\varepsilon>^{1/3}\, r^{-2/3}$, but only a fraction can cause drop breakup. Such vigorous eddies have α values below α', which can be found from Eq. (13)

$$\alpha' = \frac{2.5\log\{L/d^0_{\max}\}}{\log\{L/d\}} - 1.5 \tag{24}$$

The fraction of sufficiently active eddies is then

$$F(\alpha \le \alpha') = \int_{0.12}^{\alpha'} P(\alpha)\, d\alpha \tag{25}$$

which may be evaluated using the distribution in Eq. (17). Finally the breakup frequency, k_b, is given by

$$k_b \sim X\langle\varepsilon\rangle^{1/3} d^{-2/3} \int_{0.12}^{\alpha'} \left(\frac{d}{L}\right)^{d_s - f(\alpha)} d\alpha \tag{26}$$

This derivation is in some ways similar to an earlier analysis[11] of the effect of a distribution of the relative velocities over a given distance, d_{max}. The multifractal description of the distributed characteristics of the inertial sub-range should be superior to the assumed Gaussian distribution of v_r. It should be noted that Eq. (26) contains an influence of scale through L.

Maintaining geometric similarity and constant mean energy dissipation rate, $<\varepsilon>$, drops of a particular size formed faster as the tank size increased[11]. An interpretation and a two-zone model have been given[11]. Turbulent intermittency, expressed by Eq. (26), offers an alternative explanation. For example for the following values: $<\varepsilon>$ = 1.6 m^2 s^{-3}, T_1 = 0.128 m, L_1 = 6.4 mm, T_2 = 0.30 m, L_2 = 15 mm, d = 0.2 mm, where the suffices 1 and 2 refer to small and large scales, Eq. (24) gave $\alpha_1' = 0.23$ and $\alpha_2' = 0.38$. This signifies that in the large tank less energetic eddies could also break drops. From Eq. (25) the cumulative fractions of dispersive eddies were $F_1\,(\alpha \le 0.23) = 2.6\times10^{-4}$ and $F_2\,(\alpha < 0.38) = 7.2\times10^{-4}$. This implies that nearly three times more eddies were effective in the larger tank, which is a possible reason why drop breakup occurred faster in this tank.

Conclusions

The widely used theory of drop breakup in the inertial sub-range of turbulent flow, leading to an exponent of –0.6 on the Weber Number, ignores intermittency and employs the time-averaged energy dissipation rate. This and other turbulence characteristics exhibit, however, distributions, which may be represented using multifractals. Rare, but violent bursts influence drop sizes and, given sufficient time, the exponent on We may be as small as –0.93. Values in the range –0.93 to –0.61 are predicted using multifractals and are consistent with experimental results. Maintaining geometric similarity and keeping the time-averaged energy dissipation rate constant, consideration of intermittency using multifractals introduces a scale-dependency into various properties of dispersed drops, which is absent in the classical approach, e.g. the final drop size should be smaller in a larger tank. A more unified theory of drop breakup, flocculation etc seems to be attainable when intermittency is included. Further work in this direction, including careful comparison with experiments, is believed to be worthwhile.

Nomenclature

a	=	exponent	[-]
d	=	drop diameter	[m]
d_{32}	=	Sauter mean drop size	[m]
d_{max}	=	maximum drop size	[m]
d^0_{max}	=	value of dmax when neglecting intermittency	[m]
d_s	=	space dimension	[-]
D	=	impeller diameter	[m]
D_0	=	constant	[-]
F	=	cumulative probability	[-]
$f(\alpha)$	=	function of α	[-]
k_b	=	breakage frequency of drops of size d	[s^{-1}]
L	=	integral scale of turbulence	[m]
N	=	impeller speed	[s^{-1}]
P	=	pressure	[Pa]
$P(\alpha)$	=	probability density function of α	[-]
r	=	distance between two points in turbulent flow	[m]
t	=	time	[s]
T	=	tank diameter	[m]
v	=	velocity	[$m \cdot s^{-1}$]
v_L	=	velocity difference over a distance L	[$m \cdot s^{-1}$]
v_r	=	velocity difference over a distance r	[$m \cdot s^{-1}$]
X	=	volume fraction of zone where breakup occurs	[-]
Re	=	impeller Reynolds Number ($N D^2 \rho/\mu$)	[-]
We	=	Weber Number for stirred tank ($N^2 D^3 \rho/\sigma$)	[-]
α	=	multifractal exponent	[-]
ε	=	turbulent energy dissipation rate per unit mass	[$m^2 \cdot s^{-3}$]
$<\varepsilon>$	=	time-average of ε over whole domain	[$m^2 \cdot s^{-3}$]
ε_r	=	average of ε over domain of size r	[$m^2 \cdot s^{-3}$]
λ	=	scale factor	[-]
μ	=	continuous phase viscosity	[$kg \cdot m^{-1} \cdot s^{-1}$]
ρ	=	continuous phase density	[$kg \cdot m^{-3}$]
σ	=	interfacial tension	[$kg \cdot s^{-2}$]
τ	=	stress	[$kg \cdot m^{-1} \cdot s^{-2}$]

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