The Stable distribution as a physically meaningful description of the size distribution from Rayleigh resonance jet break up type atomisers

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Abstract

During the development of an atomiser working on the principle of Rayleigh resonance jet break up for industrial scale spray drying, it was found that the narrow droplet size distribution from this "Acoustic Atomiser" was inadequately described by the distribution functions commonly used for sprays, but well described by the Stable distribution. The alpha parameter of this distribution was found to tend towards the Gaussian limit for low viscosity fluids and the Lorentz limit with increasing viscosity, consistent with behaviour as a simple and damped forced harmonic oscillator respectively, and hence with the physics of the atomisation process.

Introduction

A surprisingly dramatic difference in spray-dried product quality has been observed [1-2] when using an atomiser which exploits the Raleigh laminar jet resonance instability [3] to produce droplets which are all of substantially the same size. A residence time scaling analysis has been used to rationalise why the product becomes so much more uniform [4-5]. The droplet size distribution model used must be a compelling fit to the data, as the analysis relies on behaviour in the 95^{th}-99^{th} percentile tail of the droplet size distribution.

Materials and Methods

The distribution functions screened include those used for sprays analysis in the literature, either commonly (log-normal and Rosin-Rammler), or rarely (hyperbolic [6-7]), and also functions not seen in the sprays literature but with a potentially appropriate peak shape (Laplace, Lorentz/Stable, log-logistic/Burr). The Stable distributions are a class of wide-tailed distributions whose instances include the Gaussian and Lorentz distributions [8]. Stable densities and cumulative function were calculated using the "STABLE" program [9].

The forty droplet size distribution datasets that were fitted were measured variously by Phase Doppler Anemometry, image analysis and sieve sizing. Mean sizes vary over two orders of magnitude: flowrate over four orders of magnitude. A few measurements are of liquid droplets, but most are of solid particle product, incorporating dispersion and artefacts in particle size due to morphology and size changes during solidification. Whilst the datasets are thus individually rather noisy, we can be confident in the robustness of a fit to such a range of data. The sources and artefacts are described in the full paper: the complete data can be found in [5].

If an observed secondary peak at small sizes were due to either satellite droplet formation or orifice size variation, then it arose from the same atomisation mechanism as the primary peak, and a compelling size distribution function should fit both peaks with meaningfully related parameters. An important property of the Stable distribution is self-similarity: if it really is a good description of the physics then the same (α, β, γ) should fit both peaks, with only the location parameter delta varying. The parameters of the hyperbolic distribution have less intuitive physical meanings and it is unclear how they should be related between peaks. Fitting to the bimodal datasets was used as a demanding test to resolve which distribution function is the best model for the data. Tertiary peaks at large diameter were attributed to agglomeration. The agglomeration process is independent from the atomisation, so self-similarity with the primary atomisation peak is not expected: it is not even clear that it should follow the same type of distribution. Thus multi-mode analysis has been confined to bimodal primary mode atomisation and the secondary mode at smaller diameter.

To evaluate the goodness-of-fit of the distribution functions to the data, in addition to the familiar volume density and cumulative volume fraction plots a delta stabilised probability (DSP) plot was used, where a data transformation makes the variance more uniform across the distribution, and hence a more sensitive test of fit to the tails than a standard probability-probability plot [8, 10]. Visual assessment of goodness-of-fit is further enhanced in the DSP plot by plotting the data so that the perfect fit line is y=0 and the 95% confidence limit acceptance bounds are horizontal. The goodness-of-fit statistic DSP [10] depends only on the sample size, so quantitative comparisons can easily be made between fits of different distribution functions to the same data.

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Results and Discussion

The log-normal, log-logistic and Burr distributions were not good fits to the data. The Rosin-Rammler was very poor. The promising fits to some of the datasets of the computationally simple Laplace and Lorentz distributions indicated that it was worth considering the complexities of the more computationally complex generalised forms of the 4-parameter log-hyperbolic and Stable distributions respectively. Both these were found to fit the data reasonably. The fitting of the hyperbolic distribution was found to be more stable than the literature [7] would suggest, but the parameters were undesirably sensitive to the selection of the data range, no pattern was apparent in the values of the parameters between datasets, and the least squares fit error in δ was found to be up to 2δ. The Stable distribution fits were for almost all datasets found to be better both qualitatively and quantitatively, and the fit parameters were always less arbitrary, especially when applied to the bimodal distribution cases when the assumption of self-similarity of the distribution between the primary atomisation and secondary satellite droplet peaks works consistently for the Stable distribution, but erratically for the hyperbolic distribution. For all datasets the agreement of the Stable distribution model with the data was found to be acceptable at the 95% confidence level, and in most cases was a comfortably small fraction of the limit.

A physical rationalisation can be made for the observed decrease in the Stable alpha parameter with increasing liquid viscosity in these resonance jet break-up nozzles. The Lorentz distribution function (Stable $\alpha = 1$) is the solution in the frequency domain of the equations of motion for a damped harmonic oscillator excited by a resonant sinusoidal fluctuation. This system is highly underdamped (otherwise the initial perturbation would decay, rather than being amplified along the jet until droplets broke off). As the viscosity (and hence the damping) increases, the system becomes progressively less like an undamped simple harmonic oscillator, and more like an ideal damped harmonic oscillator.

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References

[1] Fiannaca, C., Threlfall-Holmes, P., WO 2005/058473 (2005).
[2] Threlfall-Holmes, P., *Solids drying in theory and practice*, IChemE Particle Technology Subject Group Meeting, UCL, 2 April 2008.
[3] Rayleigh, *Proc. Royal Society* 29:71-97 (1879).
[4] Threlfall-Holmes, P., Fiannaca, C., Oliver, R., *19th Annual meeting of ILASS (Europe)*, Nottingham, UK, 6-8 September, 2004, p. K3.
[5] Threlfall-Holmes, P., PhD thesis, Heriot-Watt University, 2009.
[6] Stanton, D. W., Senecal, P. K., Hung, C. C., Rutland, C. J., Reitz, R. D., *Atomization and Sprays* 8:363-392 (1998).
[7] Xu, T.-H., Durst, F., Tropea, C., *Atomization and Sprays* 3:109-124 (1993).
[8] Nolan, J. P., *Stable distributions - models for heavy tailed data*, Birkhäuser, 2009.
[9] Nolan, J. P., *Commun. Statist. - Stochastic Models* 13:759-774 (1997).
[10] Michael, J. R., *Biometrika* 70:11-17 (1983).

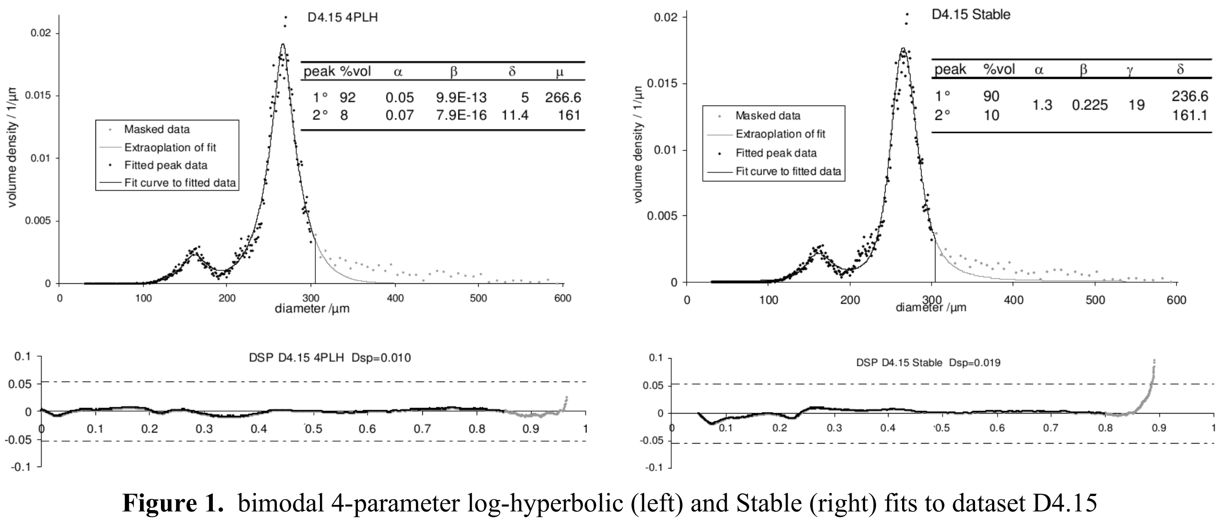

Figure 1. bimodal 4-parameter log-hyperbolic (left) and Stable (right) fits to dataset D4.15