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Short communication

A modified Nukiyama–Tanasawa distribution function and a Rosin–Rammler model for the particle-size-distribution analysis

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Abstract

No fundamental mechanism or model enables a theory on particle-size distribution to be built. Consequently, a wide variety of empirical models or equations have been proposed to characterize experimental particle-size distributions, such as the Rosin–Rammler model. Because the Nukiyama–Tanasawa equation uses four parameters to simulate differential distribution frequencies for particle-size diameters, the distribution function is not easy to apply in order to fit experimental data. In this paper, a modification of the Nukiyama–Tanasawa model with only two parameters has been proposed to fit the data on a particle-size distribution (PSD). The proposed normalized distribution function has been applied successfully to the PSD analysis (cork granulate and spray atomization droplets).

Keywords: Particle size; Distribution function

1. Introduction

Particle-size distribution is probably the most important physical characteristic of solids. This property influences the combustion efficiency of pulverized coal, the setting time of cements, the flow characteristics of granular materials, the compacting and sintering behaviour of metallurgical powders, etc. These examples illustrate the important role of particle size in energy generation, industrial processes, and many other phenomena. Also, particle size is a significant parameter in some Unit Operations such as spray drying, fluidization engineering or fluid–solid heterogeneous reactions.

Many distribution functions have been proposed to characterize the fraction of materials as a function of the particle size [1–3]. These models can be applied to describe particle-size distribution (PSD). Usually, the equations use two parameters, one representing the mean diameter and the other one indicating the particle-size range. The Rosin–Rammler distribution function, Eq. (1), is the most commonly used equation for PSD analysis.

$$F_M(D) = 1 - \exp\left(-\left(\frac{D}{m}\right)^n\right) \tag{1}$$

This function is suitable to characterize powders obtained from grinding, milling, and crushing operations [4] and to analyse the PSD of spray droplets [2,3]. However, the use of the Nukiyama–Tanasawa equation, Eq. (2), is difficult because four parameters are required: K_D, m, n and p [5,6].

$$f_{\mathrm{N}}(D) = K_D D^p \exp\left(-\left(\frac{D}{m}\right)^n\right) \tag{2}$$

The present work analyses the Rosin–Rammler and Nukiyama–Tanasawa distribution functions and proposes a more practical version of Nukiyama–Tanasawa function that use the Sauter mean diameter, D_{VS}, and two parameters, n and p.

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2. Theory

2.1. Distribution functions for particle-size distribution

The particle-number fraction between particle sizes D and $D+\Delta D$ is $f_N(D)\ \Delta D$, where $f_N(D)$ is a density function (its dimensions have to be length^{-1}). The sieve analysis of powder materials implies that the frequency distribution has to be determined using mass fractions [5]. In this case, the density function based on mass fractions is $f_M(D)$ and $f_M(D)\ \Delta D$ is the mass fraction for a particle-size range between D and $D+\Delta D$. Both distribution functions must satisfy the normalization conditions:

$$\int_0^\infty f_N(D)dD = 1; \quad \text{and} \quad \int_0^\infty f_M(D)dD = 1 \tag{3}$$

Assuming that all particles have equal shape and density:

$$f_M(D) = \frac{D^3 f_N(D)}{\int_0^\infty D^3 f_N(D)dD} \tag{4}$$

2.2. Modified Nukiyama–Tanasawa and Rosin–Rammler distribution functions

Differential forms of the Rosin–Rammler distribution function are shown below. Eq. (5) has been formulated by differentiation of Eq. (1), while Eq. (6) may be derived from Eq. (3) and the correlation between $f_M(D)$ and $f_N(D)$, Eq. (4).

$$f_M(D) = \frac{n}{m}\left(\frac{D}{m}\right)^{n-1} \exp\left(-\left(\frac{D}{m}\right)^n\right) \tag{5}$$

$$f_N(D) = \frac{n}{m\Gamma\left(\frac{n-3}{n}\right)}\left(\frac{D}{m}\right)^{n-4} \exp\left(-\left(\frac{D}{m}\right)^n\right) \tag{6}$$

The mathematical transformation is indicated in Appendix A. The Rosin–Rammler distribution functions, Eqs. (5) and (6), require two parameters to fit the experimental data, m and n.

The Nukiyama–Tanasawa equation, Eq. (2), apparently depends on four parameters (K_D, m, n and p). Lefebvre [2] or Li and Tankin [7] assume that $p=2$. In the mathematical methodology proposed, if the normalization conditions, Eq. (3), must be satisfied, it follows that (see Appendix A):

$$K_D = \frac{n}{m^{p+1}\Gamma\left(\frac{p+1}{p}\right)} \tag{7}$$

Substituting Eq. (7) into Eq. (2) gives a Nukiyama–Tanasawa equation with three parameters: m, n and p, Eq. (8).

$$f_N(D) = \frac{n\mathrm{D}^p}{m^{p+1}\Gamma\left(\frac{p+1}{n}\right)} \exp\left(-\left(\frac{D}{m}\right)^n\right) \tag{8}$$

However, a Nukiyama–Tanasawa equation useful for mass fraction data, Eq. (9), has been formulated using Eqs. (4) and (8).

$$f_M(D) = \frac{nD^{p+3}}{m^{p+4}\Gamma\left(\frac{p+4}{n}\right)} \exp\left(-\left(\frac{D}{m}\right)^n\right) \tag{9}$$

Some authors, Lefebvre [2] and Dunbar and Hickey [5], also use $p=2$ in Eq. (9), obtaining Eq. (10).

$$f_M(D) = \frac{nD^5}{m^6\Gamma\left(\frac{6}{n}\right)} \exp\left(-\left(\frac{D}{m}\right)^n\right) \tag{10}$$

The Nukiyama–Tanasawa equations, Eqs. (8) and (9), can be rearranged in a dimensionless form in order to make them easier to apply without any previous value assignment to any parameter. For that purpose, the Sauter mean diameter (D_{VS}) based on particle volume/surface ratio is defined as

$$D_{VS} = \frac{\int_0^\infty D^3 f_N(D)dD}{\int_0^\infty D^2 f_N(D)dD} \tag{11}$$

Substitution of Eq. (8) into Eq. (11) and integration gives an equation to calculate the Sauter mean diameter from the Nukiyama–Tanasawa equation:

$$D_{VS} = \frac{m\Gamma\left(\frac{p+4}{n}\right)}{\Gamma\left(\frac{p+3}{n}\right)} \tag{12}$$

However, a dimensionless diameter, δ, and a normalized distribution frequency, $f_N(\delta)$, have been defined:

$$\delta = \frac{D}{D_{VS}}; \quad dD = D_{VS}d\delta \tag{13}$$

The frequency distribution, $f_N(\delta)$, calculated as a function of δ must fulfill the normalization conditions

$$\int_0^\infty f_N(D)dD = \int_0^\infty f_N(\delta)d\delta = \int_0^\infty f_N(\delta)\frac{dD}{D_{VS}} = 1 \tag{14}$$

and therefore

$$f_N(\delta) = D_{VS} f_N(D) \tag{15}$$

Substituting δ instead D in Eq. (8), the Nukiyama and Tanasawa equation can be written in normalized and dimensionless form as:

$$f_N(\delta) = \frac{n}{\Gamma\left(\frac{p+1}{n}\right)}\left(\frac{\Gamma\left(\frac{p+4}{n}\right)}{\Gamma\left(\frac{p+3}{n}\right)}\right)^{p+1} \delta^p \exp\left(-\left(\frac{\Gamma\left(\frac{p+4}{n}\right)}{\Gamma\left(\frac{p+3}{n}\right)}\right)^n \delta^n\right) \tag{16}$$

Eq. (16) has only two parameters, n and p, making it possible to fit the experimental data with ordinary calculating programs.

The Nukiyama–Tanasawa distribution function based on mass fractions can be determined assuming that $f_M(D)$ and $f_M(\delta)$ fulfill the normalization conditions, Eq. (3), and therefore

$$f_M(\delta) = D_{VS} f_M(D) \tag{17}$$

Taking into account Eqs. (12), (13), and (17), Eq. (9) can be transformed into:

$$f_M(\delta) = \frac{n}{\Gamma\left(\frac{p+4}{n}\right)}\left(\frac{\Gamma\left(\frac{p+4}{n}\right)}{\Gamma\left(\frac{p+3}{n}\right)}\right)^{p+4} \delta^{p+3} \exp\left(-\left(\frac{\Gamma\left(\frac{p+4}{n}\right)}{\Gamma\left(\frac{p+3}{n}\right)}\right)^n \delta^n\right) \tag{18}$$

When the variable change in Eqs. (16) and (18) is undone, Nukiyama–Tanasawa equations with two parameters are obtained, Eqs. (19) and (20). These equations are as easy to use as the Rosin–Rammler equations, given that these distribution functions have only two parameters because D_{VS} can be numerically calculated from the experimental data using Eq. (11).

$$f_N(D) = \frac{n}{D_{VS}\Gamma\left(\frac{p+1}{n}\right)}\left(\frac{\Gamma\left(\frac{p+4}{n}\right)}{\Gamma\left(\frac{p+3}{n}\right)}\right)^{p+1} \times\left(\frac{D}{D_{VS}}\right)^{p}\exp\left(-\left(\frac{\Gamma\left(\frac{p+4}{n}\right)}{\Gamma\left(\frac{p+3}{n}\right)}\right)^{n}\left(\frac{D}{D_{VS}}\right)^{n}\right) \tag{19}$$

$$f_M(D) = \frac{n}{D_{VS}\Gamma\left(\frac{p+4}{n}\right)}\left(\frac{\Gamma\left(\frac{p+4}{n}\right)}{\Gamma\left(\frac{p+3}{n}\right)}\right)^{p+4} \times\left(\frac{D}{D_{VS}}\right)^{p+3}\exp\left(-\left(\frac{\Gamma\left(\frac{p+4}{n}\right)}{\Gamma\left(\frac{p+3}{n}\right)}\right)^{n}\left(\frac{D}{D_{VS}}\right)^{n}\right) \tag{20}$$

3. Results and discussion

For verification of the goodness of the fit of the Nukiyama–Tanasawa and Rosin–Rammler equations, experimental droplet-size distributions obtained by atomization were used [3,7], together with the data on cork granulates obtained by milling [4]. Data from Masters [3] and Li and Tankin [7] were used to calculate the normalized frequency of size distribution, f_N (D), by Eq. (21):

$$f_N(D) = \frac{N}{\int_0^\infty NdD} = \frac{x_N}{\int_0^\infty x_N dD} \tag{21}$$

where x_N is the droplet fraction with a size between D and $D+\Delta D$. The values obtained for f_N (D) from Masters [3] are shown in Fig. 1. The Sauter mean diameter was calculated by numerical integration of Eq. (11). Data from Macías-García et al.

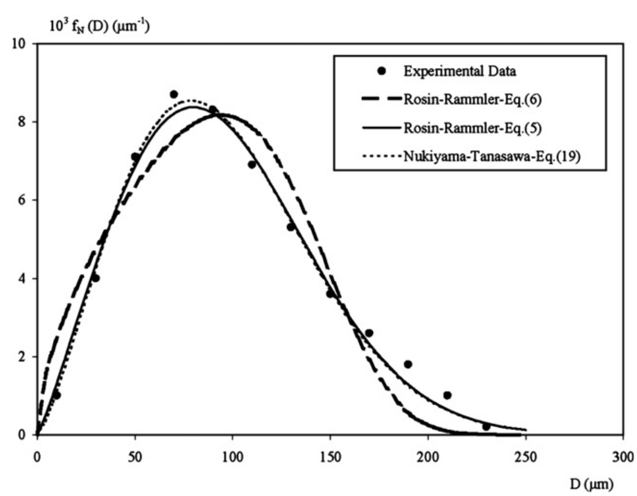

Fig. 1. Fit of the Rosin–Rammler and modified Nukiyama–Tanasawa equations to experimental data from Masters [3].

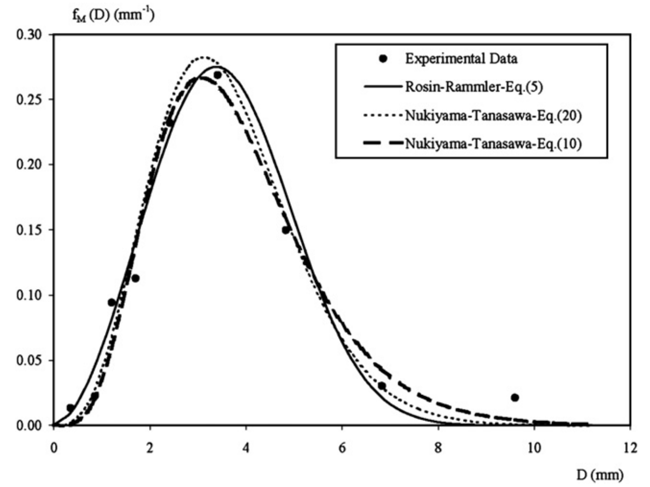

Fig. 2. Fit of the Rosin–Rammler and modified Nukiyama–Tanasawa equations to experimental data from Macías-García et al. [4].

[4] were used to calculate mass fraction of solids, x_M and f_M (D) for a particle size between D and $D+\Delta D$ and were shown in Fig. 2. From these data, it is possible to establish the mass fractions and the sieve diameters as the arithmetic mean of two sieve openings of two consecutive meshes. Similar to Eq. (21):

$$f_M(D) = \frac{x_M}{\int_0^\infty x_M dD} \tag{22}$$

The definition of the Sauter mean diameter, Eq. (11), and the correlation between f_N (D) and f_M (D), Eq. (4), leads to an

Table 1
Rosin–Rammler and modified Nukiyama–Tanasawa equations parameters

Equation	m	n	p	RSR	AMAD (%)
Masters (2001) [3] data	(μm)			10^8	
Rosin–Rammler Eq. (5)	107.36	2.14		9.810	0.021
Rosin–Rammler Eq. (6)	147.33	4.59		82.74	0.185
Nukiyama–Tanasawa Eq. (19)		1.97	1.29	7.018	0.017
Li and Tankin [7] data	(μm)			10^8	
Rosin–Rammler Eq. (5)					
E-1	168.57	2.08		4.190	0.137
E-2	164.25	2.36		22.43	0.154
E-3	151.12	2.48		5.358	0.200
E-4	169.96	2.36		14.44	0.280
Nukiyama–Tanasawa Eq. (19)					
E-1		1.83	1.14	4.871	0.022
E-2		2.04	1.29	25.39	0.029
E-3		2.07	1.29	14.26	0.042
E-4		1.83	1.13	28.11	0.012
Macías-García et al. [4] data	(mm)			10^4	
Rosin–Rammler Eq. (5)	3.982	2.76		3.269	0.82
Nukiyama–Tanasawa Eq. (20)		1.50	0.185	3.362	2.24
Nukiyama–Tanasawa Eq. (10)	0.442	0.89	2	2.997	2.35

E-1, E-2, E-3 and E-4 correspond to different experiments on droplet-size distributions measured by [7].
AMAD = Absolute mean average deviation = $\sum_0^{NB}\left|\frac{f(D)_{exp}-f(D)_{calc}}{f(D)_{exp}}\right|\frac{1}{NB}$.

expression to calculate the Sauter mean diameters as a function of sieve-analysis data

$$D_{\mathrm{VS}} = \frac{\int_0^\infty f_{\mathrm{M}}(D)dD}{\int_0^\infty \frac{f_{\mathrm{M}}(D)dD}{D}} \tag{23}$$

Matlab 6.5 was used to fit the data series. The program minimizes the sum of the residual squares, RSR, for each particle-size distribution

$$\mathrm{RSR} = \sum_1^n \left(f_{\mathrm{N}}(D)_{\mathrm{calc}} - f_{\mathrm{N}}(D)_{\mathrm{exp}}\right)^2 / \mathrm{NB} \tag{24}$$

where NB is the number of experimental data. For f_{M} (D), RSR is calculated substituting f_{M} (D) in Eq. (24) instead of f_{N} (D).

Table 1 shows the parameters n, m or p values obtained when the Rosin–Rammler and Nukiyama–Tanasawa equations are used. The RSR, the absolute mean average deviation (AMAD), and Figs. 1 and 2 show that both the Rosin–Rammler and modified Nukiyama–Tanasawa equations acceptably fit the experimental data (Table 1). The largest deviations between experimental and calculated results have been found using the modified Rosin–Rammler equation, Eq. (6). However, in order to calculate particle-size distributions, the original Rosin–Rammler equation, Eq. (5), apparently has a wider application range because it acceptably reproduces experimental results using particle-number fraction data instead of mass fractions data (Fig. 1).

4. Conclusions

The Nukiyama–Tanasawa equation, Eq. (2), has been normalized in a dimensionless form to be used in PSD analysis. The mathematical models used to study the PSD analysis have been applied to two different samples: cork granulates and atomization sprays. The modified Nukiyama–Tanasawa distribution function with only two parameters provides good results when it is applied to the samples studied. This function is easily used with common calculating tools. This means that the proposed distribution function is as useful as the well-known Rosin–Rammler equation.

Appendix A

Some calculation examples are shown. Functions have been normalized including the gamma function:

$$\Gamma(x) = \int_0^\infty y^{x-1}\exp(-y)dy \tag{A-1}$$

Normalization of the Nukiyama–Tanasawa equation

Substituting Eq. (2) into Eq. (3) leads to

$$\int_0^\infty K_D D^p \exp\left(-\left(\frac{D}{m}\right)^n\right)dD = 1 \tag{A-2}$$

Making the following variable change:

$$y = \left(\frac{D}{m}\right)^n; \quad D = my^{\frac{1}{n}}; \quad dD = \frac{m}{n}y^{\frac{1-n}{n}}dy \tag{A-3}$$

Substituting Eq. (A-3) into Eq. (A-2) and operating

$$K_D \int_0^\infty \frac{m^{p+1}}{n} y^{\frac{p+1-n}{n}} \exp(-y)dy = 1 \tag{A-4}$$

and comparing Eqs. (A-1) and (A-4), gives

$$x - 1 = \frac{p+1-n}{n}; \quad x = \frac{p+1}{n} \tag{A-5}$$

$$K_D \Gamma\left(\frac{p+1}{n}\right)\frac{m^{p+1}}{n} = 1 \tag{A-6}$$

and consequently

$$K_D = \frac{n}{m^{p+1}\Gamma\left(\frac{p+1}{n}\right)} \tag{A-7}$$

Rosin–Rammler modified equation (f_{N} (D)—Eq. (5))

Considering the correlation between f_{N} (D) and f_{M} (D), Eq. (4), and the normalized frequency distribution data, established by Eq. (3), the result from Eq. (4) is

$$f_{\mathrm{N}}(D) = \frac{C \cdot f_{\mathrm{M}}(D)}{D^3} \tag{A-8}$$

where C is a constant. Substituting Eq. (5) into Eq. (A-8) and including the resulting equation into Eq. (3)

$$\int_0^\infty C\frac{n}{m}D^{-3}\left(\frac{D}{m}\right)^{n-1} \exp\left(-\left(\frac{D}{m}\right)^n\right)dD = 1 \tag{A-9}$$

When a procedure analogous to the previous one is followed to obtain the gamma function, Eqs. (A-3) and (A-4), the following is obtained

$$C = \frac{m^3}{\Gamma\left(\frac{n-3}{n}\right)} \tag{A-10}$$

where Eq. (A-10) allows the calculation of the Rosin–Rammler equation based on particles or droplet fractions as,

$$f_{\mathrm{N}}(D) = \frac{n}{m\Gamma\left(\frac{n-3}{n}\right)}\left(\frac{D}{m}\right)^{n-4} \exp\left(-\left(\frac{D}{m}\right)^n\right) \tag{A-11}$$

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