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1. Introduction.

The aim of this paper is to offer a possible solution of a particular problem of great practical interest in the large field of particle size analysis (dimensional analysis of homogeneous ground powders in dilute suspensions), which we have studied in the last few years [1-3].

Ground powder particle size analysis is an important problem for many purposes, and light scattering by small particles is the main method generally used to solve it. In most cases the practical interest powders contain particles several times larger than light wavelength, and the Fraunhofer diffraction theory imposes itself as mathematical basis [4], due both to its
simplicity and to the fact that it leads to numerical algorithms which do not require tedious calibration procedures.

The object of all these algorithms is to compute the volumetric size distribution \( V(a) \) \( (a \) being the particle diameter), using a discrete measured data vector:

\[
e = e_n(r_{int}, r_{ext}), \quad n = 1, \ldots, N
\]

(where \( r_{int}, r_{ext} \) are the internal and external radii of each of the \( N \) ring-shaped detecting cells — almost all detectors developed for such measurements are either concentric annular systems or concentric annular sectors, due to the geometry of scattered radiation), i.e. to invert a first kind Fredholm integral equation:

\[
e_n = \int_{a_{min}}^{a_{max}} K(a, r_{int}, r_{ext}) V(a) \, da, \quad n = 1, \ldots, N
\]

where \( a_{min}, a_{max} \) are the extreme diameters of the distribution and \( K \) is the Bessel functions based kernel of the integral equation.


One of the newest and most powerful techniques dedicated to solving inverse problems is the singular value decomposition. Since this method is presented in detail elsewhere [1, 5, 6], we shall remind here only that it is largely based [7] on the use of a window coefficients set, \( W_{\mu, i} \), dependent on a regularization parameter, \( \mu \), computed by means of the input data vector, \( e \).

Beside the already mentioned advantages [1] of this method, we must underline here its great flexibility: many related algorithms may be easily developed on the basis of various distribution functions used in particle size analysis. While a new window coefficients set using Gauss distribution has been previously tested [8], we are working here with the interesting Rosin-Rammler distribution function.

This semi-empirical distribution was derived from some considerations concerning the grinding of coal [9], and showed to be — somehow unexpectedly — very adequate for ground powders characterization. The differential form of the volume distribution is:

\[
\frac{1}{V_1} \frac{dV(a)}{da} = bna^{n-1} e^{-ba^n}
\]

with \( V_1 \) = the total volume of particles, \( b \) and \( n \) = positive constants. The cumulative form of the Rosin-Rammler distribution may also be used:

\[
\frac{V(a)}{V_1} = 1 - e^{-ba^n}
\]

Due to the large acceptance of the sieving method as a reference method, equation (4) is usually written in the form:

\[
\frac{V'(> a)}{V_1} = e^{-ba^n}
\]

where \( V'(> a)/V_1 \) is denoted by \( R \) and signifies the refuse on the sieve of mesh « \( a \) ».
For practical applications the constant $b$ is replaced by a characteristic diameter, $a'$, and equation (5) becomes:

$$R = e^{-\left(\frac{a}{a'}\right)^n}$$

(6)

This equation (used in industry as Rosin-Rammler-Sperling or Rosin-Rammler-Bennet diagrams) is more convenient since the characteristic diameter $a'$ has a clearer physical meaning than the parameter $b$: $a'$ is the diameter for which $R = e^{-1}$.

The parameter $n$ is called the uniformity index and it is related to the general aspect of the distribution function, being larger for narrow distributions. For most ground powders it usually lies between 0.7 and 1.5.

3. Results and discussion.

3.1 Experimental device. — The apparatus we have used is a rather classical one (Fig. 1), with a single He-Ne laser and with a 16-cell hybrid structure detector (12 concentric ring sectors centred on the optical axis and 4 external rectangular cells) [1]. We must stress the particular importance of the simultaneous measurement of a large collection of particles: while in other samples (glass microspheres, colloidal suspensions, biological suspensions and so on) the scattering particles are almost spherical, the particles resulting from grinding processes have highly irregular shapes; that is why the expander (item 2, Fig. 1), beside its role in optical filtering the incident light, increases the beam diameter up to 20 mm. The sample undergoes an ultrasonic deflocculation, in order to prevent particles aggregation.

![Optical design of the experimental arrangement](image)

Fig. 1. — Optical design of the experimental arrangement: 1: 1.5 mW He-Ne laser, 2: Beam expander, 3: Measuring cell, 4: Pump, 5: Collecting lens, 6: Photodetector matrix.

3.2 Influence parameters. — Without any claim of being exhaustive, we worked on four types of Rosin-Rammler window coefficients:

a) RR1:

$$W_{\mu, k} = e^{-\left(\frac{\bar{a}_k}{a'_k}\right)^n}\mu$$

(7)

where $\bar{a}_k, k = 1, \ldots, N$ are the mean diameters of the granulometric classes;

b) RR2:

$$W_{\mu, k} = e^{-\left[\frac{\bar{a}_k}{a'_k} \left(\frac{\lambda_k}{\lambda_1}\right)^{\frac{1}{2}}\right]^n}\mu$$

(8)
c) RR3:

\[ W_{\mu, k} = e^{-\mu \left( \frac{a}{a'} \right)^n \frac{\lambda_k^2 - \mu}{\lambda_k^2}} \quad (9) \]

d) RR4:

\[ W_{\mu, k} = e^{-\mu \left( \frac{a_k}{a'} \right)^n \frac{\lambda_k + \mu}{\lambda_k}} \quad (10) \]

As in [1], we chose \( N' = N = 16 \), \( a_{mn} = 2 \mu m \), \( a_{mx} = 496 \mu m \), \( \lambda_0 = 6.328 \AA \) (He-Ne laser), the focal length of the collecting lens (item 5, Fig. 1) \( f = 200 \) mm, precalculated data for the singular values \( \lambda_k \) and singular vectors \( v_k \) and a precision of 0.001 for the computation of the regularization parameter, \( \mu \).

Beside the signal-to-noise ratio (S/N), which is the single critical parameter for singular algorithms using other windows quoted in literature, two more parameters are used by the Rosin-Rammler window: the characteristic diameter, \( a' \), and the uniformity index, \( n \). If the uniformity index presents small variations (typically two or three tenths for samples of the same ground powder but very different in size), however the characteristic diameter is obviously unknown.

We solved this problem by an iterative procedure: the program firstly runs with \( a' = 248 \mu m \) (the middle of the measuring range) and on the basis of the computed distribution function it founds the new value of the characteristic diameter; this value is used to compute again the distribution function, and the routine continues until two successive values of \( a' \) differ by less than 1 \( \mu m \), which we consider to be a reasonably low value.

Two important facts support this approach to the problem:

- for all the tests carried out, the values of the regularization parameter are much smaller than the same values computed with other windows [1], and consequently the speed of the program is high;
- fortunately, the procedure exhibits a fast convergence. As a matter of fact, this was one of the points in selecting a particular form of the window coefficients among the equations (7-10); only RR1, RR3 and RR4 were fast enough (for any kind of data vectors considered the needed precision in \( a' \) computing is reached in maximum five steps).

RR3 window led to unsatisfactory results for small particles and also presents a quite high sensitivity with respect to the S/N ratio.

The last criterion used to choose the actual window coefficients was the influence of the uniformity index, \( n \), on the results. Finally we have decided for RR4 window, which is practically insensitive to variations of \( n \) in the range \([0.75, 2]\) and to variations of S/N ratio in the range \([0.1, 0.2]\), being at the same time more accurate in maxima positioning than RR1.

### 3.3 Maxima Positioning Precision Tests

For an easier comparison we ran these tests in the same conditions as presented in [1]. We worked with input data errors of 20 % (S/N ratio of 5, respectively) and with \( n = 1.5 \); these values were preserved for all the other tests. The input data vectors were 16 simulated \( \delta \)-Dirac functions, centred at the middle of each granulometric class. The results (computed as relative deviations) presented in figure 2 for Tikhonov (Tk) window (probably the best window currently quoted in literature) and RR4 window, clearly emphasize a better behaviour of the last one, especially for small particle diameters, where some problems normally appear, caused by the limited validity of the Fraunhofer diffraction approximation at such dimensions.
Nevertheless, the RR window is not so successfully with respect to the broadening effect of narrow distributions. The FWHM of the computed distributions for δ-Dirac data vectors are about 1.2-1.4 times greater than those obtained with Tk window.

3.4 Maxima values tests. — As in [1], we ran our algorithm with RR4 window for several δ-Dirac input data vectors covering a granulometric class which ranges from 31.04 to 43.9 μm. The results, for Tk window and RR4 window, are shown in figure 3. While for data vectors centred close to the middle of the granulometric class Tk window yields greater percentage than RR window, for data vectors located near the class limits RR window produces larger values than Tk window.

Also, the use of RR window removes maximum shift toward large diameters, and leads to smaller variations of the computed percentage with respect to the data vector location in that class. The other tests performed with simulated data — which are likewise those presented in [1] — mark better results obtained with Tk window for δ-Dirac data vectors (mainly a greater sensitivity for low percentage local maxima) but also some advantages of RR window for Gauss-type data vectors.

3.5 Real data tests. — Since Rosin-Rammler distribution was developed to deal with ground powders analysis and on the other hand our particular interest in particle size analysis has the same direction, we focused our attention on such samples.

3.5.1 Talcum powder test. — As we had no information on the size distribution of our talcum powder, we chose the optical microscopy as a reference method — despite the high technologies involved in particle sizing, we believe that the microscopical method still remains the most reliable one. The observations made render evident the irregular shape of many particles, thus emphasizing the need for a simultaneous measurement of a great number of particles.

Fig. 2. — Relative deviations of computed maxima positions for RR window (continuous line) and Tk window (dashed line) vs. particle diameter.
Fig. 3. — Percentage values in a granulometric class for RR window (continuous line) and Tk window (dashed line) vs. δ-Dirac data vector location in the same class.

The histogram of this powder (Fig. 4) was obtained by microscopically measuring 5000 particles, and it has a typical appearance for ground powders, without narrow and well-separated maxima.

The cumulative curves of the microscopically determined distribution function and of the distribution functions computed with Tk and RR windows respectively, are presented in

Fig. 4. — Microscopically determined histogram of the talcum sample.
The very good agreement between the microscopically determined curve and the one computed with RR window is obvious.

![Cumulative curves for the talcum sample](image)

**Figure 5.** Cumulative curves for the talcum sample as computed with RR window (continuous line), Tk window (dashed line) and microscopically (dotted line) respectively.

### 3.5.2 Cement powder test.

We used a national standard cement powder, whose cumulative curve was determined on a statistical basis from measurements made with many different methods, including sieving, optical microscopy and sedimentation technique. A good agreement exists among this curve and the cumulative curve of the sample, as computed with RR window (Fig. 6).

The larger error appearing at small diameters \(a < 5 \mu m\) is due both to the measuring range (the lower limit of our device and algorithm is 2 \(\mu m\) and we have no information on the distribution below this limit) and to the failure of the theoretical model at small dimensions.

### 3.5.3 Copper ore powder test.

A typical sample — as it results from flotation technologies — was used. Its cumulative curve was computed from data obtained by sieving, the standard method of mining industry. The plots presented in figure 7 show a very good agreement between the cumulative curve as computed with RR window and that obtained by sieving.

Two observations, common to these real data tests, must be made:

- we used throughout the cumulative curves since this is the only way to compare distributions obtained by various methods when the granulometric classes are not coincident; more than that, the cumulative curve is widely used in practice;
- the use of Tk window exhibits an interesting «collapsing» effect of the computed distribution: the maximum is enhanced to the detriment of the tails of the distribution, both at small and at large diameters. Although this effect is not very stressed, it is real, may be observed for all samples and modifies the distribution.
Fig. 6. — Cumulative curves for the cement sample as computed with RR window (continuous line), Tk window (dashed line) and standard (dotted line) respectively.

Fig. 7. — Cumulative curves for the copper ore sample as computed with RR window (continuous line), Tk window (dashed line) and by sieving (dotted line) respectively.


The new Rosin-Rammler coefficients introduced here turned out to be very fruitful for the special application they were developed for (ground powders size analysis). The simulated data tests show that the use of these window coefficients preserves the general advantages of
the singular value decomposition algorithm: presents very good resolution and numerical stability, admits large input data errors, has a good resolving power of local maxima and correctly identifies small amplitude maxima.

The real data tests emphasize that the Rosin-Rammler window is more adequate for ground powders analysis while other windows quoted in literature (Tikhonov window in particular) are suitable for strongly asymmetric distributions, with narrow and well-separated maxima, but do not fit so well ground powders distributions.

The comparison made between the results obtained with the Rosin-Rammler window and with other methods currently used in particle size analysis (microscopy, sieving, sedimentation) points out in most cases a remarkably good agreement.

References

[8] Dimofte C., Mihut L. and Baltog I., « Gauss Window for Singular System Analysis in Granulometry », submitted to the 4th Int. Conf. on Optics ROMOPTO’94.