

AIR IONS AND ELECTRICAL AEROSOL ANALYSIS

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The measuring block of a granulometer or an electric analyser of aerosols gives direct information about the particle size spectrum in the form of a set of channel signals $\psi = \{\psi_1, \psi_2, \dots, \psi_n\}$. The particle size spectrum is also described by a set of numbers $\varphi = \{\varphi_1, \varphi_2, \dots, \varphi_n\}$ (e.g. fraction concentrations of particles). For a measurement method based on the primitive mathematical model of the measurement process, the estimate of the spectrum is obtained through the scale transformation

$$\psi_j = a_j \varphi_j \quad (1)$$

and graduation is reduced to determining the constants of a_j .

If a modified primitive mathematical model is used, then a preliminary transformation of the primary channel signals ψ' to secondary ψ'' is admissible (e.g. using the formula $\psi_j'' = \psi_j' - \psi_{j-1}'$). This mathematical model is used as the data processing basis of the well-known device *TSI-3030*, however, the use of the primitive model sets strict requirements to the design of the granulometer, whereas these requirements can be met only approximately.

According to the general linear mathematical model the channel signals are dependent on the size spectrum according to the linear apparatus equation $\psi_j = \sum_k H_{jk} \varphi_k$ or

$$\psi = H\varphi, \quad (2)$$

where the table of coefficients H is called the apparatus matrix.

The size spectrum of particles is calculated by solving the apparatus equation using the least squares method.

The general linear mathematical model excludes strict requirements to the equipment and makes it possible to avoid simplifications and approximations which cannot be avoided in the case of the primitive mathematical model. The primitive model is a special case of the general model and it demands a diagonal structure of the apparatus matrix. If the general linear mathematical model is used, then the graduation of the granulometer is reduced to the determination of the elements

of the apparatus matrix H_{jk} .

For the graduation of the granulometer information from various sources may be used. We will consider four methods of graduation different in the kind of information used: absolute, etalon aerosols, analysis of aerosol deposit, and complex methods.

In absolute graduation the granulometer is viewed as an absolute measuring device which is graduated by theoretical calculation on the basis of the measurement of geometrical and electrical parameters of the device. A general outline of absolute graduation is presented in [1].

The apparatus matrix is calculated according to the formula

$$H_{jk} = \frac{e}{\int_{r_{k-1}}^{r_k} f^{(k)}(r) dr} \int_{r_{k-1}}^{r_k} f^{(k)}(r) \sum_{\gamma} \gamma P_{\gamma}(r) G_j(k_{\gamma}(r)) dr, \quad (3)$$

where e - the elementary charge; G_j - the apparatus function of the measuring capacitor; index j - the channel number; $P_{\gamma}(r)$ - the probability of getting into the capacitor of particles with the size r and charge γe ; $k_{\gamma}(r)$ - the mobility of a γ -times charged particle calculated by the Stokes-Millikan formula.

In formula (3) for the sake of simplicity the fraction model of the particle size spectrum has been adopted

$$f(r) = \sum_k \varphi_k f^{(k)}(r) / \int_{r_{k-1}}^{r_k} f^{(k)}(r) dr, \quad (4)$$

whereas the function $f^{(k)}(r)$ is considered to be defined, and the experiment is to determine only the value of φ . If under the integral sign in equation (3) the function is smooth and the fractions narrow, then the apparatus matrix is practically independent of the concrete shape of the function $f^{(k)}(r)$ and those functions can be chosen arbitrarily. In the general case the values of φ_k are viewed as the coordinates of linear expansion of the particle size spectrum by elementary spectra $f^{(k)}(r)$

$$f(r) = \sum_k \varphi_k f^{(k)}(r). \quad (5)$$

Electric aerosol granulometer can be viewed as an absolute device only if the function $P_{\gamma}(r)$ is known. In practice, the

function of particle charging $P_{\gamma}(r)$ is sufficiently well known only in the case of a steady bipolar particle charging. The latter gives the devices with a bipolar charging a certain advantage. However, those devices are significantly less sensitive than devices with unipolar particle charging.

The research in the charging of aerosol particles shows that the charge distribution of particles depends on the field strength E_0 and the quantity $kn_0 t$ in the charging zone, where n_0 - the ion concentration; k - the ion mobility; and t - the time the particles stay in the charging zone. The above distribution depends also on the initial distribution the influence of which decreases with the increase of $kn_0 t$. Let us denote this distribution, normed to a unity, through $P_{1\gamma}(r/kn_0 t, E_0)$. This function describes aerosol charging in an ideal charger which guarantees identical values of E_0 and $kn_0 t$ for all aerosol particles. In real chargers aerosol particles pass through zones with different values of E_0 and $kn_0 t$. Particles may also be precipitated in the inlet of device and the charger. Let us denote the probability of a particle passing through the inlet of device and the charger in the zone with a known value of E_0 and $kn_0 t$ through $P_2(kn_0 t, E_0, r)$. Then

$$P_{\gamma}(r) = P_{1\gamma}(r/kn_0 t, E_0) P_2(kn_0 t, E_0, r). \quad (6)$$

If the ion flows onto a particle are known, then $P_{1\gamma}(r/kn_0 t, E_0)$ are determined by an equation proposed in [2]. Application of this formula for the range of large sizes causes substantial difficulties [3]. Therefore an approximate method for the determination of $P_{1\gamma}$ has been proposed for aerosol particles which are much larger than the mean free path of air ions [4]. In this case $P_{1\gamma}$ is described by the following equation

$$P_{1\gamma} = \frac{1}{\sigma_{\gamma} \sqrt{2\pi}} \exp\left(-\frac{x^2}{2}\right) \left\{ 1 + \frac{\nu_1}{6}(x^3 - 3x) + \frac{\nu_2}{24}(x^4 - 6x^2 + 3) + \frac{\nu_1^2}{72}(x^6 - 15x^4 + 45x^2 - 15) \right\}, \quad (7)$$

$$x = \frac{\gamma - \bar{\gamma}}{\sigma_{\gamma}},$$

where $\bar{\gamma}$ - the average particle charge, σ_{γ} - the standard de-

viation, v_1 - the coefficient of asymmetry and v_2 - the coefficient of excess. On the basis of this formula a system of four differential equations has been formulated, there dependencies of \bar{y} , σ_y , v_1 , v_2 on $kn_0 t$ are wanted. This system of differential equations was solved with the method of Runge-Kutta for diffusion charging taking into account the ion flow onto a particle (formula in [5]). The dependencies $\bar{y}(r, kn_0 t)$, $\sigma_y(r, kn_0 t)$, $v_1(r, kn_0 t)$, $v_2(r, kn_0 t)$ are represented as polynomials. Using the above methods for the determination of $P_{1y}(r/kn_0 t, E_0)$ for charging in the electric field, the ion flow onto a particle is to be determined. For this purpose it is necessary to solve the differential equation of charging which simultaneously takes into account the diffusion of ions and their directed movement in the summary electric field created by external surface charges and the charged and polarized particle itself. On the basis of numeric solution of this equation approximate formulas of the ion flow to conductive particles have been found. For instance, for $7 \leq W \leq 130$ and $0 \leq \mu \leq 2.4W + 3.87$ the following formula can be used

$$\frac{d\mu}{d\tau} = 3W \left(1 - \frac{\mu}{3W}\right)^2 + \exp \left[-4.567 \left\{ \sqrt{\left(1 - \frac{\mu}{3W}\right)^2 + 0.01159 \cdot \ln(3W) + 0.02806} + \left(\frac{\mu}{3W} - 1\right) - 0.04182 \cdot \ln(3W) + 1.198 \right\} \right] \quad (8)$$

where $W = \frac{eE_0 r}{KT}$ - the non-dimensional strength of the elec-

tric field, $\mu = \frac{q e^2}{4\pi \epsilon_0 r K T}$ - the non-dimensional charge;

$\tau = \frac{e k n_0 t}{4 \epsilon_0}$ - the nondimensional time, ϵ_0 - the electric con-

stant, T - the absolute temperature, K - the Boltzmann constant. To determine $P_{1y}(r)$ for particles with radii coinciding with or less than the mean free path of air ions, the above-mentioned equation [2] can be used and the necessary

values of the ion flow can be calculated according to the theory of Fuchs [6].

The shape of the function $P_2(kn_0 t, E_0, r)$ depends on the design of the charger and is to be determined for every particular case. Charging conditions in effective unipolar chargers are non-uniform and there are no sufficiently precise methods to establish the field distribution in a real charger. Theoretical calculations do not ensure the precision necessary for calibration, therefore in making up an apparatus matrix some free parameters connected with constructional and physical parameters of the device are left into the matrix; these parameters are to be established empirically.

Theoretically the most simple is a calibration method which is based on the use of test aerosols with spectra proportional to separate elementary spectra of expansion (5). Then an element of the apparatus matrix H_{jk} is determined directly as the signal of the j -th channel at the k -th separate elementary spectrum. However, practical difficulties occur here, connected with the generation and attestation of the test-aerosols for the whole range of particle sizes covered by the electric granulometer.

Calibration of an electric granulometer by the distribution of precipitated particles has been proposed in [7]. It is assumed that

- the precipitation coefficients of the particles of each group in the analyzer η_I were determined independently of the described procedure of calibration;
- the particles precipitated in the analyzer stick to the electrodes and could be measured with an electron or optical microscope separately in each measurement channel;
- the electric mobilities of the particles precipitated in each measurement channel are limited to certain intervals, and their means k_j^* are known.

There are two possibilities for the calibration: firstly, a polydisperse aerosol with approximately spherical particles the sizes of which cover the whole calibrated range is sucked through the granulometer, or, secondly, only an aerosol covering one sub-interval of the size range is used, and subsequently aerosols covering the next subintervals are used in a successive order. The precise size distribution of the particles may be unknown. At the stage of preliminary analysis of the calibration observations, the sizes

of the particles gathered on the electrodes of different channels are measured, then, on the basis of the initial size table, particle size spectra by channels $f_j(r)$ are calculated. These functions $f_j(r)$ may be approximated by expanding them by the elementary spectra of the adopted model of the spectrum:

$$f_j(r) = \sum_I f_{jI} f^{(I)}(r). \quad (9)$$

Information presented in the table of coefficients f_{jI} is sufficient for the calibration of the granulometer.

Elements of the apparatus matrix are calculated according to the equation

$$H_{jI} = \alpha F \xi_j \eta_I f_{jI} k_I^* \int f^{(I)}(r) dr / (k_I \sum_k f_{kI}), \quad (10)$$

where F - the aerosol flow, k_I - the average particle mobility, ξ_j - the channel sensitivities, i.e. $\psi_j = \xi_j I_j$, I_j - the current of charged aerosol particles in the j -th channel.

The most exact results are ensured by a complex calibration of the granulometer. In this case all sources of information are used, i.e. the theoretical model, the measurement results of etalon aerosols, and the analysis of the precipitation gathered on the channel electrodes. Let us consider one possible method of complex graduation. In this method the theoretical model is used, first and foremost, for the parametrization of the apparatus matrix. Let us denote the parameters of the apparatus matrix with $(h)_1, (h)_2, \dots, (h)_m$. On the basis of the results of the measurement of geometrical and electrical parameters of the measuring capacitor and the chargers, absolute estimates of the parameters of the apparatus matrix are calculated; these estimates make up the vector h_A . In order to use this particular estimate for complex graduation, it is then necessary to estimate the standard deviations of possible errors of these estimates $(\sigma_A)_j$, and also the correlation between the errors $(\varphi_A)_{jk}$. On the basis of these data a covariation matrix of absolute graduation C_A with the elements $(C_A)_{jk} = (\sigma_A)_j (\sigma_A)_k (\varphi_A)_{jk}$ is set up. The vector h_A and the matrix C_A contain the whole information about absolute calibration.

By the results of the measurement of the etalon aerosols an independent estimate is made of the parameters of the same theoretically founded model of the apparatus matrix. The result is formulated as some other vector of the estimate of

parameters h_L and as the respective covariation matrix of estimate errors C_L . The same course of action is followed in the case of the data obtained in the analysis of the precipitation of particles by channels. The respective estimate is denoted with h_d and C_d . Let us suppose that the estimate errors of the parameters of the apparatus matrix have a normal distribution, and that h_A, h_L, h_d are particular estimates of maximum probability. Then the particular functions of probability are written as

$$L_\xi(h/h_\xi) = \text{const}_\xi \exp(-\frac{1}{2}(h - h_\xi)^T C^{-1}(h - h_\xi)), \quad (11)$$

where ξ denotes the index A, L, or d. As all the information sources are independent, the joint probability function is

$$L_o = L_A L_L L_d.$$

This function is described with the vector h_o which is the most probable complex estimate of the parameters of the apparatus matrix, and the covariation matrix C_o which describes the precision of the estimate. It can be simply shown that

$$C_o = (C_A^{-1} + C_L^{-1} + C_d^{-1})^{-1}, \quad (12)$$

$$h_o = C_o (C_A^{-1} h_A + C_L^{-1} h_L + C_d^{-1} h_d).$$

The above method, despite its apparent complexity of realization guarantees maximum reliability in the graduation of electric aerosol granulometers.

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References see below.

APPENDIX

to the article translated above

As was pointed out in the article, direct empirical graduation of an electric granulometer directly on the basis of test aerosols is a technically complicated task. Despite that, a modified version of direct empirical graduation was

carried out by Mirme [8]. In Mirme's paper the spectra of the test aerosols did not coincide with the elementary spectra but were expanded by the elementary spectra. A sophisticated test aerosol generation equipment [9] made it possible to get experimental information sufficient to obtain an acceptable condition number of the equation system connecting the set of granulometer records with the known spectra of the test aerosols. This makes it possible to calculate the unknown elements of the apparatus matrix from the system:

A detailed algorithm for one method of complex graduation of a granulometer is presented in [10]. According to this algorithm the parameters of a theoretical model of the electric spectrometer are determined by the fitting of theoretically calculated quantities to the respective empirical quantities obtained in the measurement of etalon aerosols and the analysis of aerosol precipitation gathered on the channel electrodes. A necessary precondition here was the formulation of a preliminary algorithm for a quick computation of particle charge distribution in any condition of unipolar charging [4, 11, 12].

The approach to the graduation of aerosol granulometers described in the above article was also used for the analysis of the properties of the well known TSI-3030 [13, 14]. The apparatus matrix of the device was calculated according to the above-mentioned algorithm from the data of test aerosol measurement. Mathematical modeling demonstrated that traditional processing (the scale transformation method) of the TSI-3030 records of a unimodal spectrum of atmospheric aerosol leads to the appearance of two peaks. This points to a danger of considerable misinterpretations of measurement results. This danger can be avoided if the results are processed in accordance with the principles presented in the above article.

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