Assessment of the Quality of Electrically Produced Standard Aerosols

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ABSTRACT

The non-uniformity of particle charges is an important problem in electrical methods for producing standard aerosols, imposing strict limitations on the usability of the obtained standard aerosols. This paper examines the quality of the electrically separated aerosols, that is, the percent of singly charged particles among all charged particles in the separated aerosol. A model for the electrical separation process was developed and the separated aerosol quality was calculated as a function of parameters of the aerosol generation process. Theoretical results show that with particle sizes $d \gg 40$ nm, a high quality standard aerosol is difficult to obtain using the common methods. Also a non-monotonic decline in quality with increasing particle size was observed, with the separated aerosol quality having a distinct minimum around the particle diameter of 200 nm. In addition to the theoretical calculations, a small number of experiments were carried out giving results (described in the appendix), which, in general, support the theoretical findings.

Keywords: DMA; Classifier; Aerosol standard; Separation.

INTRODUCTION

Standard aerosols – aerosols with a well-determined particle size spectrum, are needed in many different areas: for scientific research in aerosol physics and chemistry, biology and medicine etc. Notably, they are essential for the calibration of aerosol measuring devices. The most convenient for use as a standard aerosol is the monodisperse (practically, quasi-monodisperse) aerosol, whose narrow size spectrum is considered to be fully characterized by three parameters: the particle mean (median, modal) diameter, some characteristic of the width of the spectrum, and the particle (number, volume, mass) concentration.

For standard aerosol generation the most well-known methods used are: the pneumatic pulverization of the aqueous suspension of the monodispersed polymer (latex) nano- and microspheres (Raabe, 1968), the pulverization of non-volatile liquids or solutions by vibrating orifice generators (Berglund and Liu, 1973), and the electrical separation of particles from a polydisperse aerosol (Liu and Pui, 1974) or from the electrospray of the solution of some large-molecule organic substances (Ude and Fernandez de la Mora, 2005). The described methods are generally limited in their usability outside specific particle size ranges, with the electrical separation method, despite its own shortcomings, possibly being considered the most versatile, as it can be more easily applied in a wide particle size range.

This paper aims to study the electrical separation method, characterizing its limitations, and examine how the quality of the standard aerosol depends on the parameters of the aerosol generation process. The study is limited to the separation from a polydisperse aerosol and as the particles generated by the methods mentioned above are spherical with their chemical composition not affecting the particle stationary charge distribution, the effects of chemical composition and geometrical shape are also not considered here. The aim of this paper is not so much of giving tools for practical use, but rather, to show in detail the need for an improvement in the currently used methods, and especially, to show why the more complicated method, proposed in (Uin *et al.*, 2009) is justified, when generating standard (calibration) aerosols.

ELECTRICAL SEPARATION

Electrical separation, developed by Liu and Pui (1974) is a method for extracting from the polydisperse aerosol (primary aerosol – PA) a narrow fraction according to the particles' electrical mobility (secondary aerosol – SA). This separation process is realized in the Hewitt-type (Hewitt, 1957) electrical classifier (Differential Mobility Analyzer – DMA). Here at least some part of the particles of PA must be charged electrically as uniformly as possible (their charge must be independent of their diameter) – then the extracted fraction of particles is narrow by the particle size as well. Uniform charging of the particles is a quite

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complicated problem. For small particles (diameter $d < 40$ nm) the problem can be solved by exposing the particles to a symmetrically bipolar ion atmosphere. There the particles obtain a stationary charge distribution, with the probability of more than one electron charge on a particle being negligible for such small particles. Now the particle mean diameter and the width of the size spectrum of SA can be found, knowing the geometric and regime parameters of the DMA (which define its transfer function); particle concentration can be determined by measuring the electric current carried by the singly charged particles to some current sensor.

In case of the cylindrical design, the ideal transfer function of the DMA, supposing nondiffusive particles, laminar air flows and ideal DMA geometry, can be described by three limiting mobilities (with the inlet and outlet flow rates being equal). The lowest mobility k_3 is the mobility of particles that started from the inner edge of the inlet and reached the ending edge of the output slit. The highest mobility k_2 is the mobility of particles that started from the outer edge of the inlet and reached the beginning edge of the outlet slit. Particles that started from the inner edge and finished at the beginning edge or started from the outer edge and finished at the ending edge, have the medium mobility k' (Hoppel, 1978; Tamm, 1992). The mobilities of separated particles fall between k_3 and k_2. The ideal transfer function has the shape of a triangle with k_3 and k_2 defining its edges and k' the center point (see also Eq. (3)).

In case of larger particles ($d > 40$ nm and especially when $d \gg 40$ nm), an aerosol with stationary charge distribution includes multiply charged particle fractions. This means that among the separated particles with a narrow mobility distribution there are multiply charged larger particles, having the same mobility as the singly charged "major" particles. As a result, SA with an asymmetric or even multimodal size distribution is obtained, which is not well suited for using as a standard aerosol (Kikas *et al.*, 1982; Reischl, 1991; Tamm, 1992). Although the effect of the multiply charged particles can be suppressed by shifting the point of separation in the particle diameter scale (see below) or by narrowing the particle charge distribution in comparison with the stationary distribution (Gupta and McMurry, 1989; Peil *et al.*, 1992), a full solution of the problem has not been achieved. The presence of multiple size fractions in SA decreases its usability as a standard aerosol and it can be said that the "quality" of the standard aerosol has decreased.

QUALITY CRITERION FOR ELECTRICALLY SEPARATED AEROSOL

To assess the quality of SA, a parameter $\xi = 0\ldots1$ (quality criterion) is defined (Uin *et al.*, 2009) as a percent of singly charged particles among all charged particles in the electrically separated aerosol (of a given charge sign):

$$\xi = \frac{n_1}{n} = \frac{n_1}{\sum\limits_{i>0} n_i},$$

(1)

where n is the total number concentration of all charged particles (of one sign) and n_1 and n_i are the number concentrations of particles with 1 and i charge(s) of the same sign, accordingly. Higher relative amount of singly charged particles in SA means higher quality. Ideally, standard aerosol should have $\xi = 1$, i.e. all charged particles are singly charged.

As PA is a fresh aerosol produced by a generator, its size distribution (density) function (size spectrum) $f(d)$ can be considered lognormal (in this paper, the size or mobility distribution of the particle number concentration is used everywhere):

$$f(d) = \frac{N}{\sqrt{2\pi} d \ln \sigma_g} \exp\left(-\frac{\ln^2 \frac{d}{d_g}}{2 \ln^2 \sigma_g} \right),$$

(2)

where d_g and σ_g are the geometric mean diameter and the geometric standard deviation, accordingly, and N is the total number concentration of particles. In case of a stationary charge distribution, the mobility distribution function $f(k)$ of PA is comprised of several components $f^i(k)$, describing i-fold charged particles (Fig. 1) (the components for negative particles were omitted).

The graph in Fig. 1 is presented in logarithmic mobility scale (for better overview), and the components of the mobility distribution function of PA are derived from a lognormal size distribution with $d_g = 200$ nm and $\sigma_g = 2.2$ (the last value is typical for pneumatic atomization generators). This gives $k^1 = 8.36\text{E-}9$ m^2/Vs (vertical axis is positioned at 0.2E-9 m^2/Vs). The curves for differently charged fractions are vertically scaled by the 1, 2 and 3-fold charging probabilities for the size distribution with d_g and σ_g given above and shifted to 2 or 3 times higher mobilities accordingly (the mobility for doubly charged particle is twice as high as the mobility of a singly charged particle of the same size). Relative half-width of the triangular DMA transfer function was taken to have a common value of 0.1 and the location of its peak $k' = 0.6k^1$. The position where $k = 0.2k^1$ is also shown, to better illustrate the results shown below in Fig. 2b.

As can be seen from Fig. 1, the different PA components overlap in the mobility scale. This means that the concentrations of differently charged particles in SA and thus its quality and overall concentration are determined by the properties of the PA distribution, i.e. the given d_g and σ_g, and by the position k' of the peak (and to a lesser degree also the width) of the DMA transfer function $H(k, k')$. With a given PA distribution, moving k' towards lower mobilities increases the SA quality while the overall SA concentration decreases.

The conventional way of generating PA – the direct charging/neutralization of big particles usually gives wide strongly overlapping differently charged PA components (PA particles usually having been produced by pneumatical atomization, giving $\sigma_g \approx 2$). It will be shown below that in these cases a commonly used simple assessment of the PA
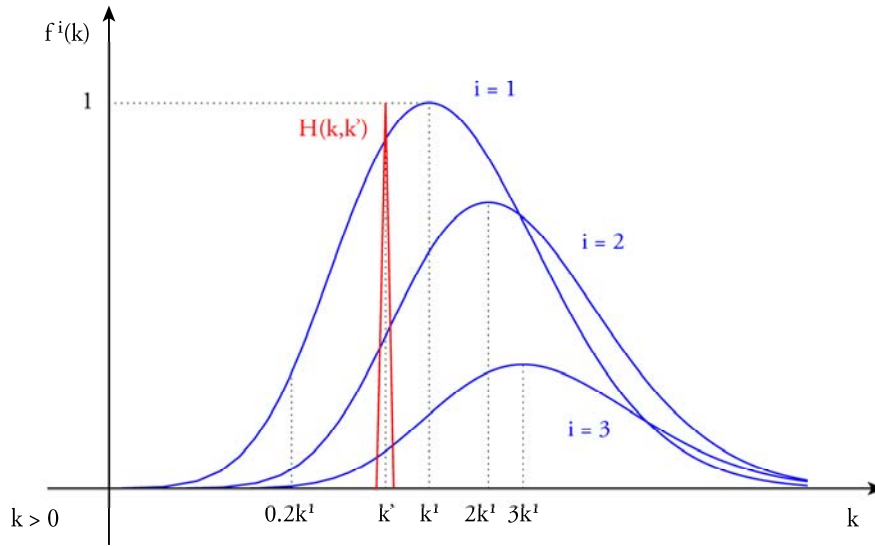
Fig. 1. Mobility distribution function of 1-, 2- and 3-fold charged components of PA and the DMA transfer function *H* in logarithmic mobility scale. All the curves are rescaled so that the singly charged component has unit height. In this case, the value of k^1 is 8.36E-9 m²/Vs and the vertical axis is positioned at 0.2E-9 m²/Vs; $k' = 0.6k^1$.

distribution for choosing the separation point (somewhere near the lower end of the PA mobility spectrum to maximize the SA quality while accepting low concentration) does not give sufficiently good results for all cases where standard aerosols are needed.

QUALITY PARAMETER CALCULATIONS FOR BIPOLAR STATIONARY CHARGE DISTRIBUTION

The objective was to calculate the quality parameter ξ as a function of the PA parameters d_g, σ_g and also find how the size region in the PA distribution where the particles are being separated from affects the SA quality. It must be noted that although bipolar charging is used, only one charge sign is considered, as the electrical separation method deals with one polarity at a time.

The transfer function *H* of the DMA was assumed to be triangular (Tammet, 1970; Stolzenburg, 1988; Stratmann *et al.*, 1997) (Fig. 1):

$$H(k,k') = \frac{\alpha}{2\beta}\left(\left|\frac{k}{k'} - (1+\beta)\right| + \left|\frac{k}{k'} - (1-\beta)\right| - 2\left|\frac{k}{k'} - 1\right|\right). \quad (3)$$

For the two transfer function parameters in Eq. (3) – the height α and relative half-width β, the actual set of values from previous experiments (Uin *et al.*, 2006) was used, however, no significant difference was observed compared to the ideal case where $\alpha = 1$ and $\beta = 0.1$ (with aerosol- to sheath air flow rate ratio of 1/10).

From the definition of the transfer function, the mobility distribution density $\varphi(k)$ of SA becomes:

$$\varphi(k) = H(k,k')f(k), \quad (4)$$

where $f(k)$ is the mobility distribution function of the PA,

normalized to the full number concentration of particles *N*. The concentration of particles in SA having *i* charges is:

$$n_i = \int_{k_3}^{k_2} H(k,k')f^i(k)\mathrm{d}k, \quad (5)$$

where $f^i(k)$ is the mobility distribution function of particles with *i* charges in PA and $k_3 = (1 - \beta)k'$ and $k_2 = (1 + \beta)k'$ are the limiting mobilities of the transfer function (see also section 2). In the particle diameter scale this can be written as

$$\frac{n_i}{N} = \int_{d(k_2)}^{d(k_3)} H(d,d(k'))f^{*i}(d)\mathrm{d}d = \int_{d(k_2)}^{d(k_3)} H(d,d(k'))p_i(d)f^*(d)\mathrm{d}d. \quad (6)$$

Here $f^*(d) = f(d)/N$ is the distribution function (Eq. (2)) of all particles and $f^{*i}(d)$ is the distribution function of particles with *i* charges in PA, both normalized to 1. The parameter $p_i(d)$ is the probability for *i* charges on particles with a given diameter; it was calculated using the Fuchs algorithm (Fuchs, 1947) with corrections by Tammet (Tammet, 1991; Hõrrak *et al.*, 1998). All the conversions between particle mobility and diameter were done using the Millikan formula.

Series of values were given to d_g and σ_g and each time the integral (Eq. (6)) was calculated for all values of *i*. The maximum value for charge number *i* was chosen to be 20 to guarantee that all significant fractions of particles were accounted for. The quality parameter ξ was calculated by its definition (Eq. (1)) and $n_N = n/N$, the relative concentration of all charged particles (of the same sign) in SA was also found. Initially the calculations were done for the situation where SA was separated from the peak of the size distribution of PA, i.e. $k' = k^1$. Next, the k' was shifted

towards the lower mobility end of the distribution and the calculations were repeated. For this a new "shift-parameter" $\gamma = 0...1$ was introduced so that $k' = \gamma k^1$.

An example of the results from these quality calculations is shown in Fig. 2. As can be seen from the first graph (a), when separating from the peak of the distribution, the SA quality decreases rapidly when particle size get higher than about 50 nm. A good quality at these larger particle sizes would be attainable only with unrealistically low values of σ_g. One method to remedy this situation has been to separate from the lower mobility end of the PA distribution, trying to avoid multiply charged particles (see Fig. 1). The second graph (Fig. 2(b)) illustrates this case. From the graph it is visible that not much improvement can actually be made this way – the quality of SA is higher, but the gain is relatively small. Also as expected, the resulting particle concentrations are much lower compared to ones obtainable when separating from the peak: the relative concentration n_N of particles in SA differs approximately five times.

The graphs in Fig. 2 also show that the general decrease in quality with an increasing d_g is not monotonic: a region of especially low quality exists in the d_g scale, after which the quality increases again, however, generally not reaching the previous maximum. This effect is especially visible on the second graph where it can be seen that this quality "valley" is located at around 200 nm. The reason for this "valley" originates from how differently charged components of PA overlap in the mobility scale which depends on the charging probabilities of particles (in Eq. (6) n_i is a function of p_i) and thus on d_g. However, the effect of charging probabilities is not examined in detail in this paper.

To give an overview of SA quality in case of different values of the PA parameters d_g and σ_g, the required shifts γ towards the lower mobility end of the distribution of PA to obtain the quality of at least 95% and the available relative particle concentrations in those cases are presented in Table 1. As can be seen, in some situations the 95% quality is unattainable with reasonable values of γ and n_N.
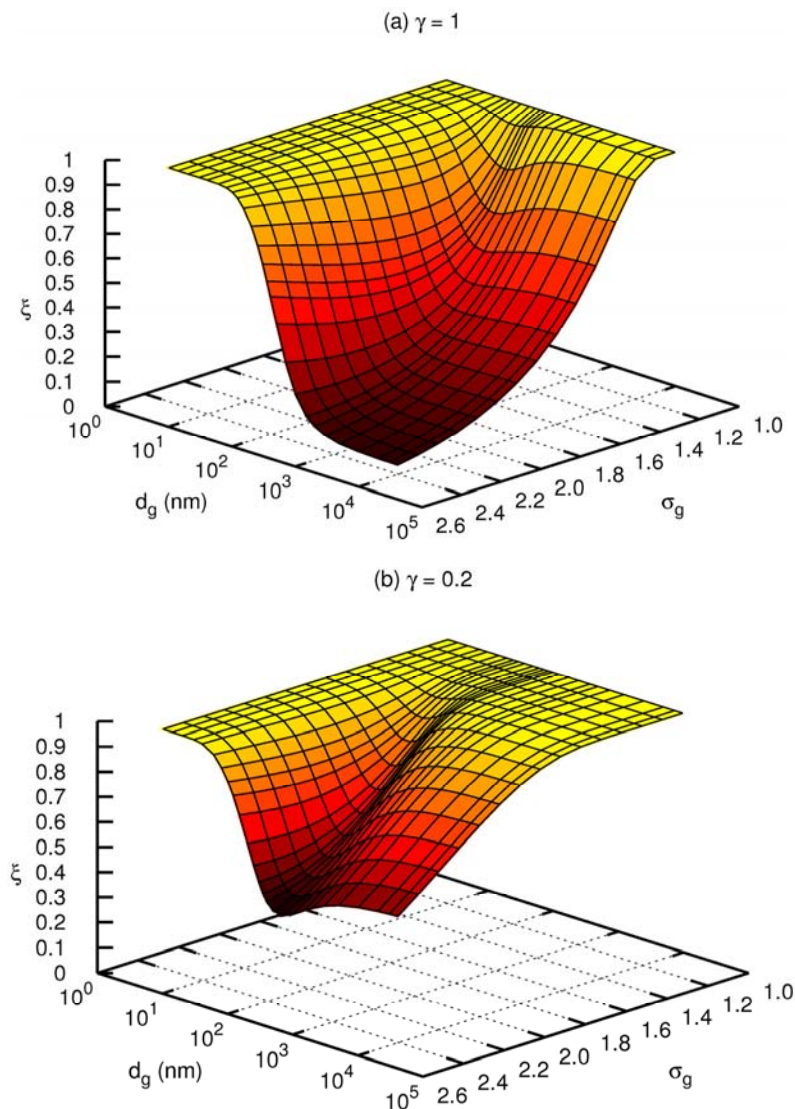
(a) $\gamma = 1$

(b) $\gamma = 0.2$

Fig. 2. SA quality parameter ξ as a function of the PA parameters d_g and σ_g. Situations where aerosol is separated from the peak (a) and from the low mobility tail (b) of the PA distribution are shown.

Table 1. Highest possible γ (smallest required shift) to obtain $\xi \geq 0.95$ and corresponding values of n_N (%) [γ; n_N] in case of different PA parameter d_g and σ_g values. N/A denotes the situation where $\gamma < 0.1$, i.e. for practical purposes the required ξ is unattainable.

σ_g	d_g (nm)				
	20	50	150	750	10000
1.2	1.00; 1.61%	1.00; 4.18%	1.00; 9.31%	1.00; 16.90%	1.00; 20.73%
1.5	1.00; 0.59%	0.18; 0.57%	0.23; 0.84%	0.31; 0.88%	0.31; 0.51%
1.8	1.00; 0.30%	N/A	N/A	0.15; 0.42%	0.18; 0.50%
2.1	1.00; 0.17%	N/A	N/A	N/A	N/A
2.4	1.00; 0.10%	N/A	N/A	N/A	N/A

As the relative amount of multiply charged particles in SA decreases when moving the separation point towards the lower mobility end of the PA (Fig. 1), beginning from a certain mobility, the concentration of doubly charged particles can be below some limit to be practically relevant/detectable (i.e. comparable in magnitude with the measurement error). However, this limit can be practically valid only for a certain instrument or setup – the fact that for a given instrument there appear to be no multiply charged particles does not mean that they are actually not present and detectable by another instrument. For some applications, accepting also low particle concentrations, such reasoning may be sufficient and the complicated calculations are not necessary. When generating standard aerosols for instrument calibration where larger particle sizes and higher concentrations are required, however, the situation is different and a guaranteed high-quality standard aerosol is required. This gives importance to the presented more thorough analysis which shows that it is not always possible to generate very good quality standard aerosols by electrical separation of bipolarly charged polydisperse aerosol. And when also high concentrations are required, a better method for producing the standard aerosols may be necessary.

CONCLUSIONS

The effect of multiply charged particles on the quality of standard aerosol produced by electrical separation was examined in this paper. It was shown that in particle size region $d \gg 40$ nm, it is quite difficult to reliably obtain a good-quality standard aerosol for instrument calibration with common methods, especially when the primary aerosol (for electrical separation) is produced using the usual pneumatic atomization and stationary charge distribution. In some applications quick estimations can be made without the presented complicated calculations, however, high quality aerosol will not be guaranteed.

An interesting effect of separated aerosol quality increasing with increasing particle sizes (starting around particle diameter of 200 nm) was also found. The effect is not very significant for practical purposes, but is worthy of further study.

The aim of this paper was to provide a more detailed analysis and illustrate the need for improvement in methods of using the electrical separation for generating standard aerosols for instrument calibration. This gives relevance to the more complicated method described in (Uin *et al.*, 2009) which completely removes the multiple charge problem and allows generating high-quality aerosols in a wide size range and with high concentrations.

NOMENCLATURE

α	height of the DMA transfer function
β	relative half-width of the DMA transfer function
γ	shift parameter, specifying the point of separation of particles
σ_g	geometric standard deviation of PA size distribution function
ξ	quality parameter for SA
φ	distribution function of the SA
d_g	particle geometric mean diameter of the PA
f	distribution function of the PA
f^*	PA distribution function, normalized to 1
f^i	PA distribution function of particles with i charges
k	electrical mobility of a charged particle
k'	midpoint mobility of the DMA transfer function
k_2, k_3	limiting mobilities of the DMA transfer function
N	total number concentration of the particles of the PA
n	total number concentration of all *charged* particles of the SA
n_1	number concentration of SA particles with single charge
n_i	number concentration of SA particles with i charges
n_N	total number concentration of all charged particles of the SA, normalized to N
p	charging probability
PA	primary aerosol (polydisperse aerosol, from which the SA is separated)
SA	secondary aerosol (quasi-monodisperse aerosol, separated from PA)

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APPENDIX. EXPERIMENTAL ASSESSMENT OF THE SA QUALITY IN CASE OF PA WITH PARTICLE BIPOLAR STATIONARY CHARGE DISTRIBUTION

A series of experiments were conducted to assess the SA quality experimentally and compare it to the results of theoretical calculations. The setup for the experiments was as follows. For aerosol generation a two-step scheme was used (Uin *et al.*, 2009) where at first silver particles (99.99% purity) with a modal diameter of approximately 18 nm were produced in a tube furnace (at temperature 1100°C) and then grown in a modified LaMer type (Sinclair and LaMer, 1949) condensational generator (growth chamber) using DOP. With this scheme, aerosols with σ_g in the range approximately from 1.28 to 1.32 (it was difficult to keep the σ_g constant) and d_g in the range of 100 to 300 nm were produced (Fig. 3). Particles were charged bipolarly with a ^{239}Pu charger and then separated with a very long Vienna-type DMA (aerosol flow rate 100 l/h, sheath air flow rate 1000 l/h) first described by Peil and Tamm (1984). As some of the particles were multiply charged, the separated aerosol consisted of differently charged particle size fractions with their mobilities in the same narrow interval.

The size distribution of the separated aerosol was then measured with an optical aerosol spectrometer (Micro Laser Particle Spectrometer from PMS), which clearly showed separate size fractions in the SA, corresponding to the differently charged particles. The particle number concentrations for these fractions were measured and the quality parameter ξ was calculated using Eq. (1). The experiment setup was then switched to mode where only singly charged particles are produced (Uin *et al.*, 2009) and the d_g and σ_g of the aerosol size distribution were found by fitting a lognormal function to the experimental data. From these values the theoretical quality parameter was calculated and compared to the one obtained experimentally (Table 2). Possible measurement errors were estimated, with the main source of error judged to be in the determination of σ_g –standard deviation from the lognormal fit (essentially possible measurement uncertainty at 67% confidence level) was ± 0.02 on average which gives approximately ± 0.03 for the theoretically calculated ξ. This also explains the varying values in the second column of the table (it was not possible to reliably obtain the same σ_g value with all experiments).

As it can be seen, there is a general agreement between the experimentally found and theoretical quality. However, the increasing trend of ξ in the d_g scale, found in the theoretical results, is very weak and (considering also the measurement uncertainties) difficult to find experimentally.
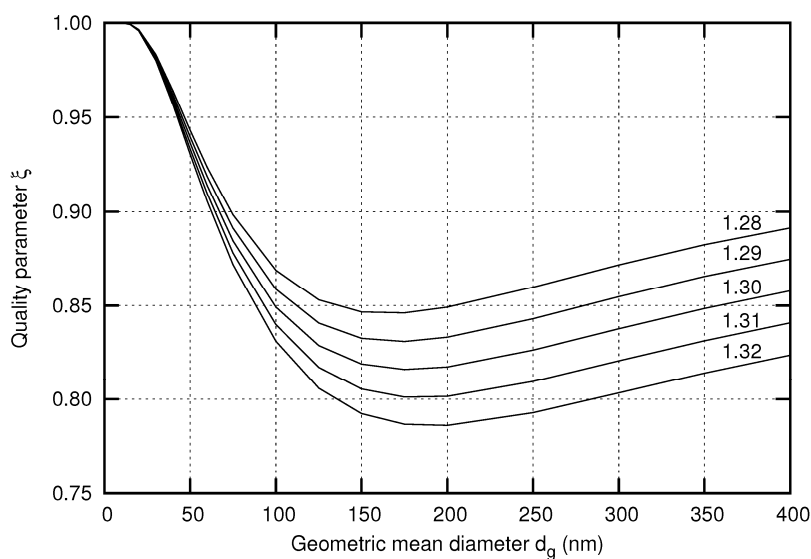
Fig. 3. Theoretical SA quality in the d_g and σ_g range used in the experiments. Different lines correspond to the different values of σ_g (shown above the corresponding line).

Table 2. Experimentally found SA quality and the corresponding calculated quality in case of the experimentally found d_g and σ_g values. Estimated measurement errors for σ_g and ξ are given in parentheses.

d_g (nm)	σ_g (± 0.02)	exp. ξ	theor. ξ (± 0.03)
156	1.29	0.86	0.83
285	1.29	0.86	0.85
138	1.31	0.81	0.81
199	1.31	0.86	0.80
363	1.31	0.90	0.83

In addition, the relatively low size resolution (in the higher diameter range) of the used optical spectrometer did not permit to reliably use particle sizes over ~375 nm, i.e., to use data points at a higher d_g where theoretically the quality should also be higher, giving the greatest possible (and more easily detectable) difference in ξ as compared to the theoretical valley. Thus, the existence of the theoretically found low-quality region remains unconfirmed in a strict sense due to the insufficient accuracy of the used instruments. However, there is evidence in favor of it as the experimental results do not show a monotonic decrease in ξ with increasing d_g which would be the case if the theoretically predicted low quality region was not present.