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A note on particle size analyses of kaolins and clays

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Abstract

Based on paradigmatic experimental results for one selected type of kaolin the systematic differences in the measurement of particle size distributions of kaolins and clays by the sedimentation method and by the low angle laser light scattering (LALLS) method are studied. A theoretically sound shape transformation procedure for sedimentation results is proposed, based on a physically justified modification of the Stokes law, which takes into account the oblate (plate-like) shape of kaolin and clay particles. It is found that, with realistic estimates of the shape factor (aspect ratio), the corrected sedimentation data come very close to the light scattering data. This indicates at the same time a way to extract shape information from the comparison of two independent size measurements. Scanning electron microscopy of the 2-μm-undersize sedimentation fraction shows kaolin particles with a disc diameter of 3–5 μm and is thus in full agreement with the interpretation of the size measure as an "equivalent disc diameter". © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Clays; Electron microscopy; Particle size analysis; Platelets

1. Introduction

Sedimentation and light scattering methods are among the most widely applied methods for particle size measurements. When identical samples are measured by either of these methods the resulting particle size distribution (PSD) may or may not be the same. To practitioners, it is well known e.g. that the systematic differences in the micron and submicron range can be considerable for clays or kaolins, whereas for micromilled sand e.g. the agreement can be quite satisfactory. A rigorous analysis reveals that the main effect has, principally, nothing to do with (and is only very indirectly influenced by) the physical limitations inherent in either method (set e.g. by the assumption of laminarity or the onset of Brownian motion in sedimentation, the validity of the assumptions for an evaluation by Fraunhofer theory etc.). The general answer to this problem is that the size measured is an equivalent diameter, i.e. the diameter of an ideal sphere which has the measured characteristic in common with the irregular and anisometric particle to be measured, and that, therefore, different methods are naturally measuring different results. This is beyond doubt. In order to enable a mutual comparison of both types of data sets — especially for purposes of industrial practice, where information about raw materials from various sources and different suppliers has to be compared, or where older (sedimentation) data bases are available and have to be compared with new (light scattering) routine measurements — many current software packages offer an option of "empirical" shape "correction" (sometimes called "revalidation"). After a sample has been measured by both methods, the PSD actually measured e.g. by light scattering is compared with the corresponding PSD measured by the other method (e.g. sedimentation), which has to be explicity fed into the computer by the operator. Results of subsequent measurements on a sample of the same or a similar type, e.g. of a new sample or a new batch can then be automatically transformed into hypothetical results which would probably have been obtained by the other method.

Although in certain limits useful from a pragmatic viewpoint, it is clear that such a procedure is totally unsatisfactory from a principal point of view. In this paper we propose a physically more justified shape transformation procedure for kaolins and clays, which can be applied for transforming classical sedimentation data (Andreasen method) with respect to low angle laser light scattering (LALLS) data and consists in the use of a modified Stokes law, which takes into account the oblate particle shape.

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In contrast to the empirical shape corrections mentioned above (which work only a posteriori, i.e. after two PSDs have been measured, one by each method), the model proposed in the present paper is the first principal step to predict (at least to a certain degree) the PSD measurable by one method (or at least certain features of it) a priori from the PSD measured by the other method and an explicit knowledge (or estimate) of the shape factor. On the other hand, when two PSDs of the same material are available, the model allows shape information to be extracted from the difference of the two PSDs.

2. Theoretical

For a general reference on theoretical and experimental aspects of particle size measurements the reader should consult Allen's standard monograph.[1] The instrumental basis of low angle laser light scattering (LALLS) is a laser beam passing through an optical cell containing a statistically representative ensemble of particles, in sufficient dilution and random orientation, and recording the spatial angular distribution of scattered light intensity by an angle-resolving photodetector. The usual evaluation procedure for the case when the wavelength of the laser light (λ) is smaller than the particle size (D), consists in a deconvolution of the scattering and interference pattern according to the integral equation:

$$I(\theta) = I(0)\int_0^\infty \left[\frac{2J_1(\alpha\sin\theta)}{\alpha\sin\theta}\right]^2 f(D)\,\mathrm{d}D \qquad (1)$$

where $I(\theta)$ is the angle-dependent light intensity directly measured experimentally, $I(0)$ the primary light intensity, $J_1(...)$ the (first kind) Bessel function, $\alpha = \pi D/\lambda$ a dimensionless size parameter, and $f(D)$ the (continuous) distribution function (frequency curve), which can be replaced by a discrete distribution for practical purposes. In lack of necessary input data (complex refractive index of the solid phase) Fraunhofer evaluation is often (especially in routine measurements) applied also in the submicron region, where, from a purely physical viewpoint, an evaluation according to the Mie theory[2] might seem more adequate. Apart from that, more fundamental considerations can speak against the use of the Mie theory: the assumption of isotropic spheres, for which the Mie theory is valid, may lead to erroneous deconvolution results for real systems with anisotropic and anisometric particles. In the present paper the equivalent diameter D calculated from LALLS measurements, using Fraunhofer evaluation (treating the particle as a black box), is called LALLS diameter. For an exact derivation of Eq. (1) and a survey of solutions of the so-called "inverse scattering problem", i.e. the extraction of information (here the "size") about the scatterer (here the particle) from the scattered light pattern, the reader may refer to standard references.[3–5] All light scattering measurements have one important feature in common: the particle size is always inferred from a projection of the particles viewed perpendicular to the laser beam direction.

Sedimentation methods on the other hand utilize the fact, that particles with different masses, i.e. also different volumes, will attain different terminal velocities when settling in viscous media. In most cases the experiment is made with dilute (in order to minimize particle–particle interactions) suspensions in sufficiently wide (in order to minimize particle–wall interactions) glass cuvettes or cylinders. For sufficiently high suspension columns and sufficiently small (i.e. slowly settling) particles the terminal velocity can be calculated from the column height h and the settling time t without significant error. The evaluation is usually performed by the Stokes formula for settling spheres, according to which the so-called Stokes diameter D_0 is

$$D_0 = \sqrt{\frac{18\eta h}{(\rho_S - \rho_L)gt}} \qquad (2)$$

where η is the viscosity coefficient (of the pure liquid medium without particles), ρ_S the density of the solid phase, ρ_L the density of the (pure) liquid phase and g the gravitational acceleration. As is well known, this equation can be derived very simply by an elementary consideration of force equilibrium, when F_R, the resistance force exerted by the viscous liquid medium on the particle,[6] is assumed to be

$$F_R = 6\pi\eta R v \qquad (3)$$

with v being the (terminal) velocity of the particle relative to the liquid medium and $R = D_0/2$ the "particle" radius (equivalent sphere radius). Apart from several assumptions of physical character (laminarity of flow, steady flow with final velocity), the validity of Eq. (2) is essentially based on the geometrical assumption that the particles are spherical. Since this is usually not the case for real systems, Eq. (2) is not "valid" in the classical sense and need not even be a good approximation to reality. The Stokes diameters D_0 calculated by Eq. (2) are diameters of hypothetical spheres with the same settling behavior as the irregular, anisometric particle in question. The Stokes diameter is, therefore, a special kind of dynamical equivalent diameter and should be considered only as one of the possible size measures derivable from sedimentation experiments. Other size measures can be extracted from sedimentation experiments, especially when something is a priori known about the particle shape. For kaolinite and many other clay minerals e.g. it can be reasonably assumed that most primary particles are pseudohexagonal platelets, which could be modelled either as oblate rotational ellipsoids (flat spheroids) or as circular discs. The exact solution for the disc case is well known from Lamb's authoritative treatise on hydrodynamics,[7] cf. also the famous textbook by Landau and Lifshitz.[8] Two extreme cases of disc orientation relative to the flow field have to be distinguished, viz. with the disc plane normal or parallel to the flow direction. The respective resistance forces are:

$$F_{\mathrm{R\ normal}} = 16\eta R v \quad (4)$$

$$F_{\mathrm{R\ parallel}} = \frac{32}{3}\eta R v \quad (5)$$

Now we define a shape factor (aspect ratio) as the ratio of disc diameter D and disc height H:

$$\Psi = \frac{D}{H} \quad (6)$$

For the case of circular discs (i.e. flat cylindrical plates) the lift force (buoyancy force) F_B and the gravitational force F_G, respectively, are calculated as follows:

$$F_B = \pi R^2 H \rho_L g = \frac{2\pi R^3}{\Psi}\rho_L g \quad (7)$$

$$F_G = \pi R^2 H \rho_S g = \frac{2\pi R^3}{\Psi}\rho_S g \quad (8)$$

Substituting for Eqs. (7) and (8), together with either Eq. (4) or Eq. (5), in the force balance yields the following disc diameters for the two extreme disc orientations, respectively:

$$D_{\mathrm{normal}} = \sqrt{\frac{32\eta h \Psi}{\pi(\rho_S - \rho_L) g t}} \quad (9)$$

$$D_{\mathrm{parallel}} = \sqrt{\frac{64\eta h \Psi}{3\pi(\rho_S - \rho_L) g t}} \quad (10)$$

The dynamical equivalent diameters thus calculated may be called equivalent disc diameters ("equivalent" because kaolin and clay platelets are not really *circular* discs), in order to distinguish them from the diameters calculated by Eq. (2), which in this connection should be called equivalent sphere diameters. For the calculation of these diameters the value of the shape factor (aspect ratio) Ψ must be known, at least approximately. Although most literature references abstain from giving concrete values for this shape factor, it seems reasonable to tentatively assume Ψ-values ranging from 10 to 30 for plate-like kaolinite and clay mineral particles and to take these values as a first approximation for kaolins and clays in which the plate-like components are prevailing.

3. Experimental

The test subject of this work is a Czech kaolin from the Pilsen basin area (type SP EX, Keramika Horní Bříza a.s., Kaznějov, Czech Republic), which is widely used in the ceramics and paper industry and whose general characteristics, including size distributions of typical sample batches, are fairly well known.[9,10] It has to be stressed, however, that the results of the present work claim to be of rather general validity for many kaolins and clays, and should principally not be dependent on any special material. The choice of kaolin SP EX has thus only paradigmatic character.

The LALLS measurements in this work were performed on the Fritsch Particle Sizer "Analysette 22" (Fritsch GmbH, Idar Oberstein, Germany), using a HeNe laser ($\lambda = 632.8$ nm) and model–independent evaluation by Fraunhofer theory. A maximum measuring range of 0.1–200 μm has been chosen. In order to exclude measuring artefacts, all adjustable parameters concerning sample treatment during measurement (volumetric flow rate, stirrer speed and ultrasonics) were kept at the same level for all measurements. Under the conditions selected the LALLS measurements turned out to be practically insensitive to previous deflocculation of the samples. Apart from the usual measurements of complete kaolin samples, individual sample fractions obtained from dried suspensions, from which particles larger than a certain limited size (corresponding to the equivalent sphere diameter determined by the sedimentation method) have already settled out, were again measured by LALLS.

The sedimentation measurements were performed by the classical Andreasen method[1] in a glass cylinder with an Andreasen pipette and a suspension column of initial height 200 mm. The progress of sedimentation is recorded by removing constant sample volumes at a known height h below the surface after a sedimentation time t. The solid phase concentration in the suspension is about 0.5 vol%. After initial tests with sodium carbonate (Na_2CO_3) an alkali-containing organic polyelectrolyte (an aqueous solution of sodium polyacrylate) has been chosen to achieve optimum deflocculation.

4. Results and discussion

Fig. 1 shows measured sedimentation data of 8 different samples of kaolin SP EX. Empty squares denote

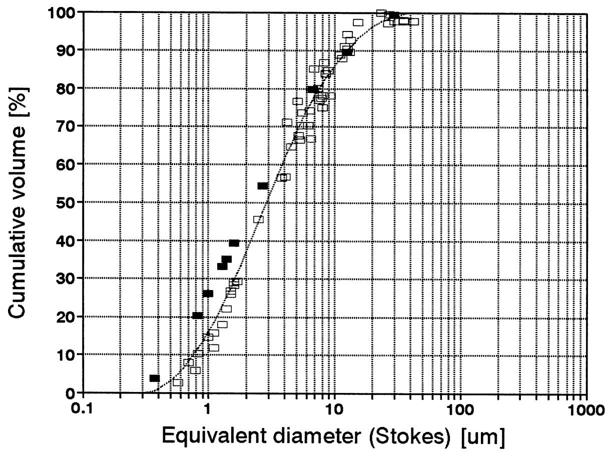

Fig. 1. Sedimentation data of kaolin SP EX (8 different samples); 7 samples deflocculated by sodium carbonate (empty squares), 1 sample deflocculated by a polyelectrolyte (filled squares), average curve fitted by nonlinear regression (dotted line).

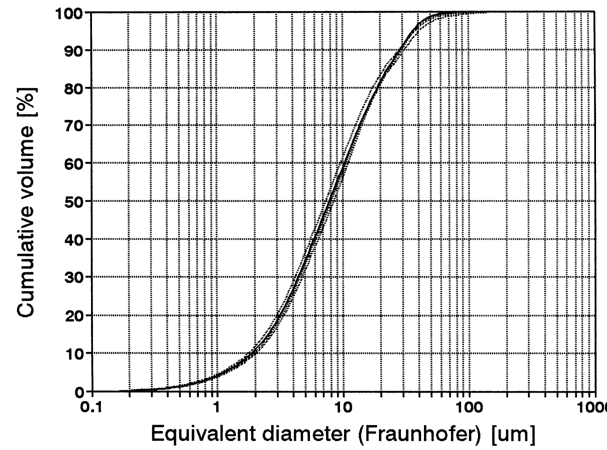

Fig. 2. LALLS data of kaolin SP EX (4 different samples); 1 sample unflocculated (dashed line), 2 samples deflocculated by sodium carbonate (dotted lines), 1 sample deflocculated by a polyelectrolyte (solid line).

data of 7 samples deflocculated by Na_2CO_3, filled squares denoting one sample (the one selected for further investigation) deflocculated by the polyelectrolyte. The relatively large scatter of the data points is due to sampling, sample preparation and statistical errors of measurement. For reasons of comparison an average curve (dotted line) has been fitted through all data points to smooth out experimental scatter and the influence of sample preparation. It is evident that, especially for particle sizes below approximately 3 μm, deflocculation by the polyelectrolyte is more efficient than deflocculation by Na_2CO_3. The average curve indicates a median size of 2.9 μm and a percentage of 37.0% below 2 μm (corresponding very well to the literature data reported in Ref. 9), while for the optimally deflocculated sample (filled squares in Fig. 1) the median size is about 2.0 μm, i.e. the percentage of particles with Stokes diameters below 2 μm is about 50.0% (in agreement with the data provided by the supplier).[10] Fig. 2 shows LALLS data of 4 different samples, one undeflocculated, two deflocculated with Na_2CO_3 and one deflocculated with the polyelectrolyte. These measurements yield a median size ranging from 7.5 to 8.7 μm, and a percentage of approximately 9.6–11.5% below 2 μm. For the sample deflocculated by the polyelectrolyte (solid line) the median size is 8.0 μm and the percentage below 2 μm (the "2-μm-undersize percentage") is 10.7%, which is well within the usual experimental scatter and indicates that — under the measuring conditions chosen — the influence of deflocculation prior to LALLS measurements is negligible for kaolin SP EX. Irrespective of all mentioned details a comparison of Figs. 1 and 2 clearly shows that the difference between the results of the two methods is a principal one and that sampling, sample preparation or statistical errors of measurement are of secondary importance. The 2-μm-undersize percentage is conventionally taken as a practically significant index for kaolin and clay characterization[11] and an "average" equivalent spherical diameter of less than 4 μm is sometimes taken as the criterion for potential plasticity.[11] It is evident that the respective values obtained by LALLS are in no way comparable to the sedimentation results (the 2-μm-undersize percentages e.g. differ by about 320–520% and the median values measured by LALLS would indicate a low degree of plasticity, which is not the case). Fig. 3 compares sedimentation and LALLS data for micromilled quartz sand, and the picture is quite different here: sedimentation and LALLS data are in satisfactory agreement. The micromilled sand particles are much more isometric than kaolin and clay particles, i.e. their (average) shape factor (aspect ratio) will be much closer to unity, although their shape is highly irregular (crushed fragments), cf. Fig. 4. This is a clear indication of the fact that the anisometry of particle shape is the main factor responsible for the large discrepancies between sedimentation and LALLS data.

In the following we confine our consideration to the one kaolin sample deflocculated with the polyelectrolyte, since the degree of dispersion achieved here can be considered as optimum. Fig. 5 compares the particle size distribution of kaolin SP EX in terms of equivalent sphere diameters [calculated from sedimentation data by the classical Stokes equation (2)] and the LALLS particle size distribution evaluated according to Fraunhofer theory.

A short note might be useful at this place: It is clear that Fraunhofer theory is but an approximation to the "exact" (in the sense of classical electrodynamics) Mie theory,[2] valid only in the case $\lambda < D$, and also it is true, that the computational difficulties connected with the Mie theory are easily managable with the computers nowadays avaliable. In practice, however, the necessary optical data for an evaluation by the Mie theory (i.e. the complex refractive index of the solid referring to the wavelength of the laser used) are often not readily accessible, and if they are, their use is highly nontrivial in the case of mixtures of components with different optical properties and in the case of optically anisotropic materials, especially when they are additionally of an anisometric particle shape, as in the case of kaolins and clays. Generally, the Mie theory is expected to affect quantitative corrections in the submicron range, i.e. for the case $\lambda \geqslant D$, when the size of the scatterer (particle) comes close to the wavelength of the probe (laser light). Fortunately, when supermicron particle sizes are present, the submicron range becomes relatively insignificant in a volume distribution for mere statistical reasons, and, therefore, in such a case eventual differences in the submicron range (due to the evaluation method chosen) will not lead to substantial changes in the overall distribution.

Irrespective of the special theory chosen for evaluation (Mie or Fraunhofer), light scattering measurements have one feature in common, as mentioned above: the particle size is always related to a projection of the particles viewed in the laser beam direction and thus, such a particle size scales with projected area. An elementary consideration for oblate particles (discs) reveals that even if we take into account the most extreme particle orientation relative to the laser beam (viz., parallel to the laser beam direction, respectively), LALLS will overestimate the statistical weight of the large-size fractions in comparison to an equivalent particle size that scales with volume or mass (as is the case of the equivalent sphere diameter calculated from sedimentation data).

Table 1 shows the "correction factors" for transforming equivalent sphere diameters into equivalent disc diameters in the case of parallel and normal orientation of the disc plane relative to the flow field, respectively. It can be seen that for a realistic range of possible shape factors ($\Psi = 10, 20, 30$) the "correction factor" for the equivalent diameters attains values between 1.94 and 4.12; the correspondingly "corrected" particle size distributions are shown on Figs. 6–8. The good overall agreement of sedimentation and LALLS results is evident, especially when shape factors Ψ of about 20–30 are chosen. These values seem realistic, although concrete

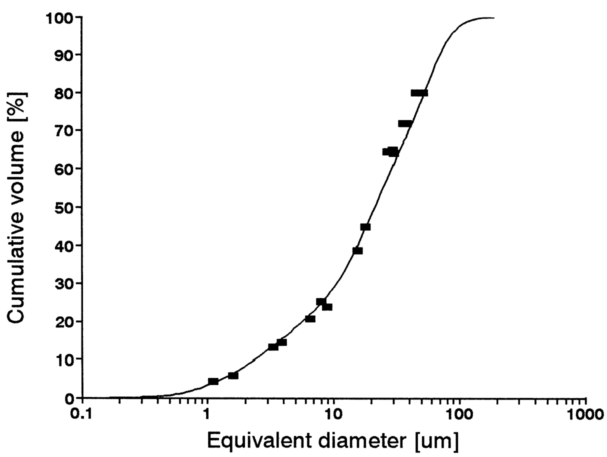

Fig. 3. Comparison of the particle size distribution of micromilled quartz sand measured by sedimentation and LALLS (filled squares and solid line, respectively).

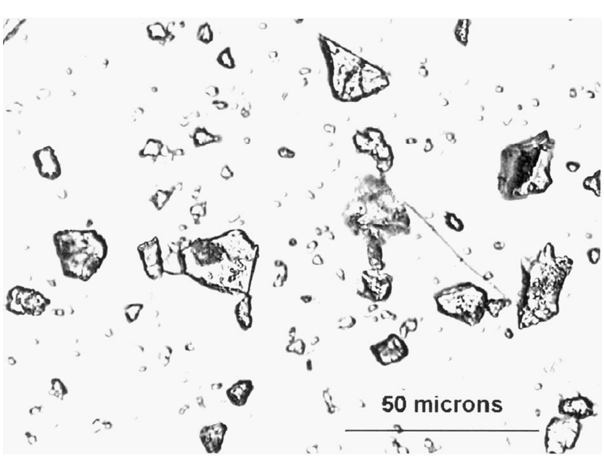

Fig. 4. SEM micrograph of micromilled quartz sand (irregular but isometric particle shape).

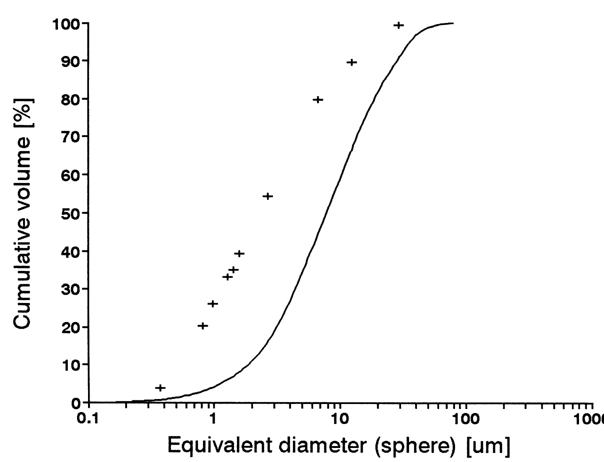

Fig. 5. Particle size distribution (sedimentation data — filled squares, LALLS curve — solid line) of kaolin SP EX deflocculated by the polyelectrolyte (sedimentation data evaluated by the classical Stokes equation for spheres).

Table 1
"Correction factors" for transforming equivalent sphere diameters into equivalent disc diameters in the case of parallel and normal orientation of the disc plane relative to the flow field, respectively (shape factors $\Psi = 10, 20, 30$)

Shape factor Ψ	Orientation parallel	Orientation normal
10	1.94	2.38
20	2.75	3.36
30	3.36	4.12

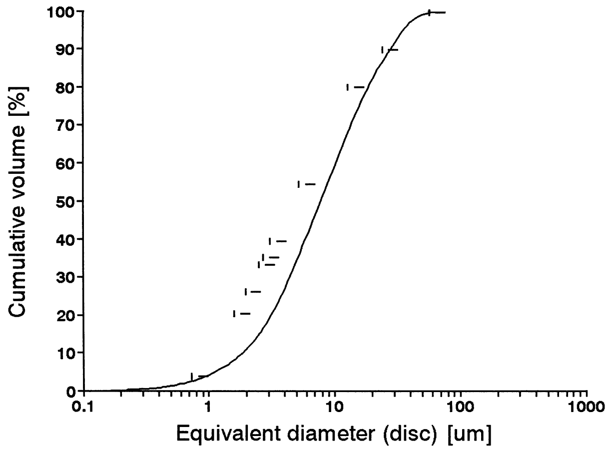

Fig. 6. Particle size distribution (sedimentation data — vertical and horizontal bars for disc parallel and disc normal, respectively, LALLS curve — solid line) of kaolin SP EX deflocculated by the polyelectrolyte (sedimentation data evaluated by the modified Stokes equations for discs with shape factor $\Psi = 10$).

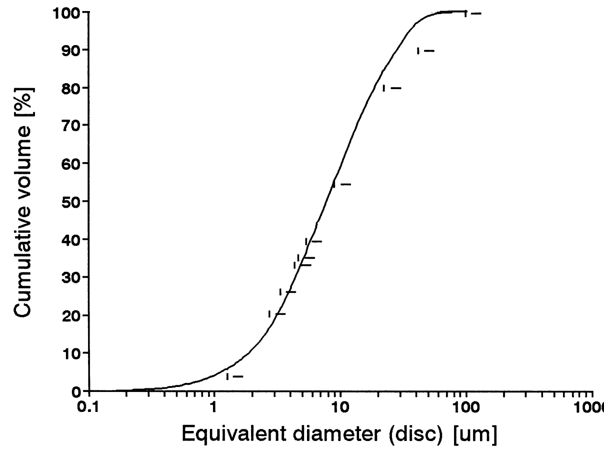

Fig. 8. Particle size distribution (sedimentation data — vertical and horizontal bars for disc parallel and disc normal, respectively, LALLS curve — solid line) of kaolin SP EX deflocculated by the polyelectrolyte (sedimentation data evaluated by the modified Stokes equations for discs with shape factor $\Psi = 30$).

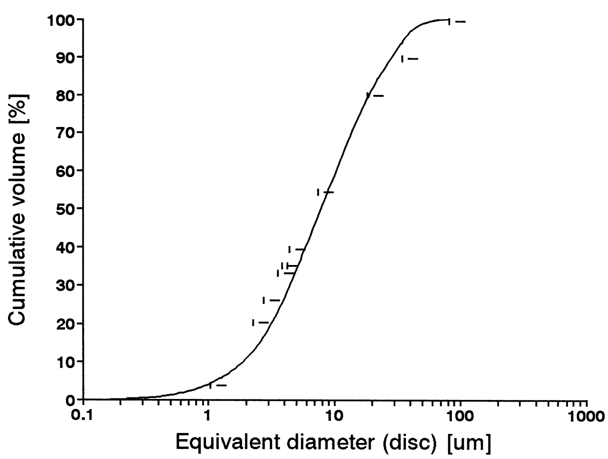

Fig. 7. Particle size distribution (sedimentation data — vertical and horizontal bars for disc parallel and disc normal, respectively, LALLS curve — solid line) of kaolin SP EX deflocculated by the polyelectrolyte (sedimentation data evaluated by the modified Stokes equations for discs with shape factor $\Psi = 20$).

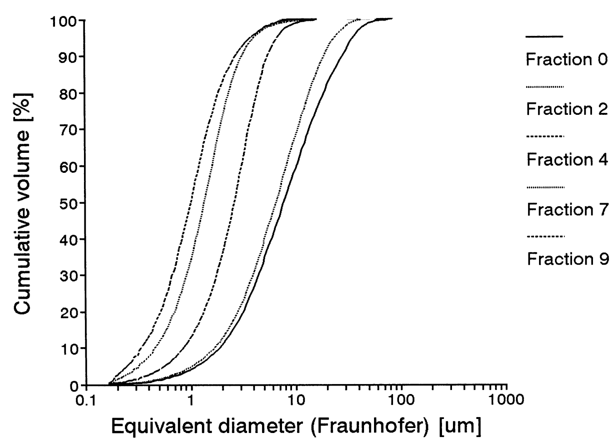

Fig. 9. Particle size distributions of selected sedimentation fractions (from right to left: 0th, 2nd, 4th, 7th and 9th fraction).

literature references for this kaolin are not available. When Fig. 8 is considered in more detail, it can be seen that the agreement is comparatively poor in the very fine size range (below 2 μm) as well as in the large size range (above 10 μm). We attribute the relatively poor coincidence in these size ranges to the fact that in both regions the shape factor is smaller than in the intermediate size range for the following reasons. In the fine size range crushed fragments of kaolin platelets prevail, while in the large size range a certain amount of agglomerates might have possibly survived. In both cases the resultant particles approach a more isometric shape and thus the shape factor will be closer to unity.

Fig. 9 shows the particle size distribution curves of five selected sedimentation fractions (from right to left 0th, 2nd, 4th, 7th and 9th fraction, cf. Table 2), while Table 2 and Fig. 10 compare sedimentation undersize diameters (equivalent sphere diameters from sedimentation measurements are always maximum equivalent sphere diameters when the undersize cumulative curve is considered) and LALLS median values for these sedimentation fractions. If sedimentation and LALLS results would yield directly comparable size measures, the LALLS median values (denoted by pluses in Fig. 10) should naturally be lower than the sedimentation undersize, i.e. maximum, diameters (dotted straight line in Fig. 10). In this case the median-versus-undersize curve should approach the dotted straight line in Fig. 10 only asymptotically. In reality, however, a crossover is observed at a sedimentation equivalent sphere diameter of about 2 μm. Principally it could be argued that this anomaly might be caused by Brownian motion (which is known to have an influence on particle

Table 2
Comparison of sedimentation undersize diameters (maximum equivalent sphere diameters) and LALLS median values for the respective sedimentation fractions

Fraction	Sedimentation undersize (μm)	Size corrected for $\Psi = 20$ (par.) (μm)	Size corrected for $\Psi = 20$ (norm.) (μm)	LALLS median (μm)
0	63 (sieve)	(173.3)	(211.7)	8.0
1	29.8	82.0	100.1	7.9
2	12.5	34.4	42.0	6.5
3	6.7	18.4	22.5	5.2
4	2.7	7.43	9.07	2.61
5	1.58	4.35	5.31	1.61
6	1.40	3.85	4.70	1.61
7	1.28	3.52	4.30	1.39
8	1.01	2.78	3.39	1.09
9	0.82	2.25	2.76	1.03
10	0.38	1.05	1.28	Unmeasurable

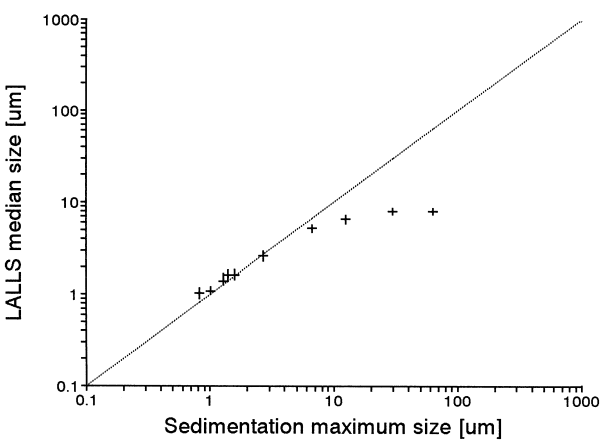

Fig. 10. LALLS median values (pluses) versus sedimentation undersize values, i.e. maximum equivalent sphere diameters; if the size measures of both methods were directly comparable, the median values should naturally be lower than the undersize values (i.e. the dotted straight line should be approached only asymptotically).

diameters of the order of magnitude 1 μm);[1,12,13] we do not, however, believe this to be the main effect in our case, because the concentration drops further from more than 40% at 2 μm (equivalent sphere diameter, i.e. after 13.6 h of sedimentation) to about 4% at 0.38 μm (i.e. after 13 days). Fig. 11 shows the same graphical representation for the case in which the modified Stokes law has been used for evaluation, cf. also Table 2. When the modified Stokes law is used, all LALLS median values are now smaller than the sedimentation undersize values (now interpreted as equivalent disc diameters). Thus, it can be concluded that sedimentation and LALLS results can be directly compared when the sedimentation data are evaluated in terms of equivalent disc diameters. Fig. 12 shows a representative SEM micrograph of the 2-μm-undersize fraction of kaolin SP EX. It is evident that particles which are assigned to the 2-μm-undersize fraction (with sedimentation data eval-

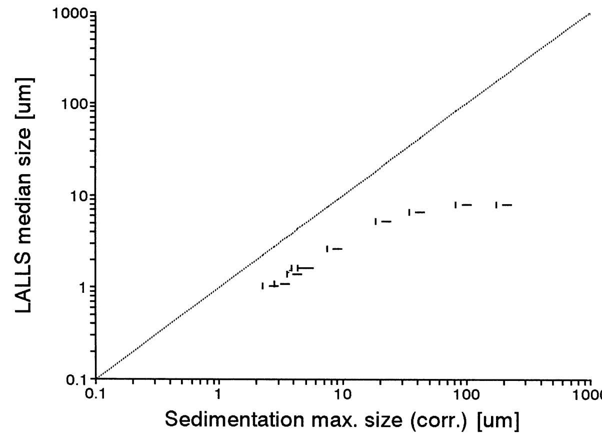

Fig. 11. LALLS median values (vertical bars for disc parallel, horizontal bars for disc normal) versus corrected sedimentation undersize values (maximum equivalent disc diameters with assumption $\Psi = 20$); the size measures of both methods are now directly comparable, median values are lower than undersize values.

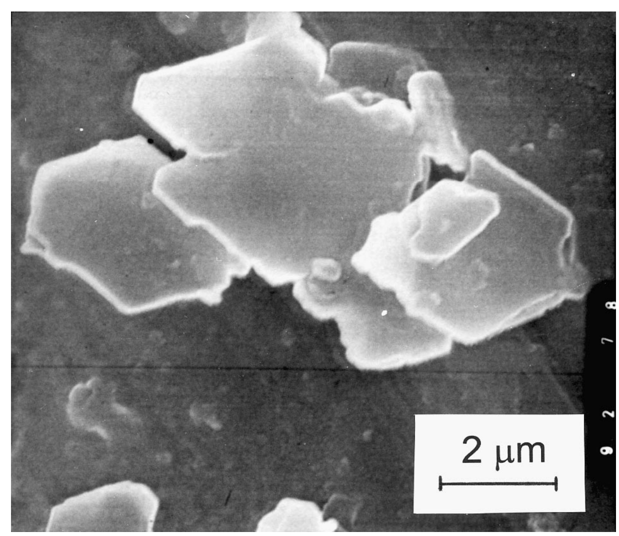

Fig. 12. SEM micrograph of the 2-μm-undersize (in terms of equivalent sphere diameters) sedimentation fraction of kaolin SP EX containing platelets with disc diameters clearly larger than 2 μm.

uated equivalent sphere diameters), exhibit (equivalent) disc diameters of 3–5 μm and larger. Far from being surprising, this finding turns out to be absolutely consistent with the above explanations and confirms the adequateness of our approach. Assuming a shape factor $\Psi = 20$, the expected equivalent disc diameter for a particle belonging to the 2-μm-undersize fraction (according to the equivalent sphere diameter) can be up to 5.5–6.7 μm. It is clear that for sieving (microsieving) the equivalent disc diameter is critical and not the equivalent sphere diameter. Thus, claims concerning the good agreement of sieve data with other particle sizing methods (which has been found e.g. by Zwicker[14,15] even for the fraction below 2 μm) that have occurred in literature from time to time[16] should not be generalized, since they cannot hold in the case of anisometric particles and are certainly invalid for kaolins and clays. This paper should have served to elucidate the relations between the relevant equivalent diameters and to emphasize the fact that not a seemingly "good agreement" between different particle size methods is desirable but an adequate interpretation of the results, for which the choice of a specially adapted evaluation of the raw data (e.g. by a modified Stokes law as proposed in the present paper) can be a helpful tool. It has not to be overlooked that the disagreement between PSDs measured by different sizing methods can be used as a source of additional information — information on particle shape.

In practice the evaluation procedure proposed in this paper for the direct comparison of sedimentation and LALLS data of kaolins and clays works due to three facts: first, the "correction factor" (from equivalent sphere diameter to equivalent disc diameter) does not differ too much for the two extreme orientations ($0.615\sqrt{\Psi}$ and $0.752\sqrt{\Psi}$ for normal and parallel orientation, respectively). Second, oblate discs of a certain minimum anisometry (the shape factor Ψ must be larger than 2.64, see above) always settle more slowly than spheres, irrespective of their orientation relative to the flow field, i.e. the "correction factor" for the corresponding equivalent diameter is in all these cases larger than unity. Third, the shape factor Ψ enters the formulae for equivalent diameter calculation (9), (10) only as a square root of Ψ and, therefore, the evaluation does not depend too critically on the exact knowledge of this shape factor. The fact that the whole particle size distribution curve is multiplied by a constant "correction factor" is of course an idealization. Therefore, we would call the proposed procedure a first-order shape transformation. A higher-order shape transformation could take into account the fact, that the shape factor Ψ varies with absolute size. Maximum shape factors are to be expected in a medium size region, where the particles (kaolin or clay platelets) are isolated but not crushed into small fragments. On the other hand, smaller shape factors can be expected for very fine particles (crushed fragments) as well as for agglomerates of particles. This problem might be solved by superposing a suitable shape factor distribution function on the PSD curve. Similarly, when shape information is to be extracted from a comparison of PSD curves measured by two different methods, a quite reasonable shape factor can of course be calculated from a comparison of the median values of the two PSD curves. It is thinkable, however, that — after taking account of all relevant fine effects (e.g. applying an appropriately modified Mie theory for LALLS evaluation in the micron and submicron region) and reducing experimental errors by optimizing sample preparation and using advanced equipment — it will be possible to obtain not only a shape factor but a shape factor distribution (SFD) curve by bringing the two PSD curves into full congruence. The SFD would show, directly, how the shape factor varies with particle size.

5. Conclusion

- The large disagreement of the results of measurements of the particle size distributions by sedimentation and by LALLS is mainly caused by deviations of the particle shape from isometry. Therefore, the results for micromilled quartz sand e.g. can show quite good agreement, whereas results for kaolins and clays are usually significantly different for the two methods.
- Satisfactory agreement between sedimentation and LALLS results for kaolins and clays can be achieved, when the sedimentation data are evaluated according to a modified Stokes law which takes into account the anisometric shape of the particles. The very simple circular disc model seems to be sufficiently flexible to serve as a basis for an adequate evaluation.
- Diameters calculated by the modified Stokes equations (9) and (10) have to be interpreted in terms of equivalent disc diameters. In order to exclude possible misunderstandings in this connection, we should like to stress once again, that no result in this paper suggests that an evaluation according to the classical Stokes Eq. (2), is wrong or as such inadequate. Interpreted in terms of equivalent sphere diameters such an evaluation is of course absolutely correct.
- Brownian motion in the sedimentation cylinder plays no significant role for the discrepancies between sedimentation and LALLS measurements; especially the large differences in the 2-μm-ndersize percentage cannot be explained by Brownian motion but must be explained by the more principal fact, that LALLS measurements yield a size measure more corresponding to equivalent disc diameters than to equivalent sphere diameters.
- Electron microscopy reveals that the 2-μm-undersize fraction (classified according to the equivalent sphere diameter), quite usually contains discs with (equivalent) disc diameters in the range 3–5 μm. This result is consistent with LALLS measurements, since these exhibit for this fraction a median (i.e. an average, not a maximum, size) of 2 μm.
- While this paper presents the possibility of a physically justified shape transformation of sedimentation data with respect to LALLS data for kaolins and clays on the basis on an a priori estimate of the shape factor Ψ, the reverse should be possible as well, i.e. a comparison of sedimentation

and LALLS data might principally be used to obtain information about the particle shape.

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