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Size and shape characterization of oblate and prolate particles

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Abstract

New methodological developments for the characterization of anisometric particles are presented and their application to real particle systems is demonstrated. In the case of oblate particles, using pyrophyllite as an example, a recently developed model relation is applied to extract approximate shape information by comparing laser diffraction and sedimentation results. The LS shape factor, an approximate measure of the average aspect ratio of the system, is found to be approx. 4.3 in the central region of the size distribution. In the case of prolate particles, taking organic crystals (mesalamine) as an example, distributions of different size measures (projected-area diameter, minimal and maximal Feret diameter) and aspect ratios are determined via microscopic image analysis and the sizing results compared with laser diffraction. It is found that the laser diffraction results exhibit a broader distribution, which is shifted in direction of the fine-size region (median diameter 16.9 μm versus 21.0 μm).

Keywords: Fibers; Platelets; Optical microscopy; Silicate; Pyrophyllite

1. Introduction

Particle size determination is a standard operation in ceramic technology and many other branches of science.[1–4] In the case of isometric particles the equivalent sphere diameter determined via various methods can be expected to be very similar, so that size distributions measured via one method should be well comparable to those measured via another.[1–4] This is not the case, however, for anisometric particles, i.e. particles with significantly different extension in different directions.

Platelet and short-fiber systems, both natural and synthetic, are very common in ceramic science and elsewhere. Platelet systems comprise the tabular forms of various oxides, nitrides and carbides (Al_2O_3, BN, SiC) as well as kaolinites and many other clay minerals (e.g. montmorillonite and hydrotalcite) and other phyllosilicates (e.g. talc). Short-fiber systems frequently used in ceramic technology include SiC (whiskers), Al_2O_3 fibers and wollastonite, but fibers are ubiquitous in other industries as well and have attracted much attention due to their potential health hazards.

In previous work various kaolin types have been characterized with respect to size and shape, using sedimentation and laser diffraction[5–8] and calculating a quantitative shape measure, the "LS shape factor" (related to an average aspect ratio of the system) via a newly proposed relation,[5,6] cf. also.[7] The characterization of wollastonite by laser diffraction has been studied in,[9] and recently two types of wollastonite with significantly different aspect ratio have been characterized via microscopic image analysis and the results compared with those of laser diffraction.[10] In the present contribution we perform a size and shape characterization of two other particle systems, which can be viewed as typical examples of oblate and prolate particle systems, respectively: pyrophyllite, a layered silicate with applications in the ceramic industry (wall tiles, refractories) and other industries, and mesalamine, fibrous organic crystals used in pharmaceutical applications. Apart from typical results, which indicate generally valid trends (irrespective of the particular systems studied), we pay special attention to quantitative error estimates, which are largely missing in the pertinent literature so far.

2. Theory

Although most anisometric particles have an irregular shape with different dimensions in all directions, many of them can approximately be considered as rotationally symmetric (e.g. cylinders or spheroids, prolate or oblate). In this case shape can be described by a single number, the aspect ratio. We define the aspect ratio R as the ratio between the maximum (D_{max}) and min-

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imum extension (D_{min}) of a particle, i.e. $R = D_{max}/D_{min}$. When this definition is adopted, $R > 1$ both for oblate and prolate particles. The case $R = 1$ corresponds to isometric or spherical shape.

In the case of oblate particles with sufficiently large aspect ratio the size distributions measured via sedimentation methods are usually shifted to finer sizes in comparison with those determined via laser diffraction. This well-known fact, together with the fact that for isometric particles the sizing results are essentially identical, can be exploited to derive a simple formula to calculate a quantity called LS shape factor, $\psi = (3\pi/4) \times (D_L/D_S)^2$, where D_L is the equivalent diameter determined via laser diffraction and D_S the equivalent diameter determined via sedimentation, cf.[5–7]. The present contribution concerns a system with a relatively small aspect ratio ($R \approx 5$), in contrast to[5–8], which deal with high-aspect-ratio particle systems ($R > 10$).

In the case of prolate particles microscopic image analysis (MIA) is clearly the method of choice. In[10] two types of wollastonite have been characterized via MIA and the results have been used as an input information for examining the suspension rheology of these systems.[11] In order to compare the (number-weighted) MIA results with the (volume-weighted) laser diffraction results a transformation procedure has been proposed and applied.[10] This procedure consists in transforming the number-weighted frequency histogram (q_0) into a volume-weighted cumulative curve (Q_3) and has been performed in[10] under the assumption that the shape, quantified via the aspect ratio R, is size-invariant. In the present contribution we investigate an example for which this is not the case. In order to take due account of the measured size dependence of the aspect ratio in the q_0–Q_3-transformation each size class measured by MIA is transformed with the individual aspect ratio of this size class.

3. Experimental

The size distribution of pyrophyllite (N.F.L. 21, nominal sieve size <45 μm, supplied by Mircal Deutschland GmbH) was characterized by X-ray sedimentation (Micromeritics Sedigraph 5100) and by laser diffraction (Fritsch Analysette 22). The sample preparation for the sedimentation measurements included homogenization and deagglomeration by high-power ultrasonics (Dr. Hielscher UP400S). Laser diffraction data (obtained with pump, stirrer and ultrasonics switched on) were evaluated using the Fraunhofer approximation. The size distribution of mesalamine (a prototype sample supplied in aqueous suspension from the pharmaceutical industry) was determined by laser diffraction (Fritsch Analysette 22) and via microscopic image analysis (Jenoptik Jenapol & Laboratory Imaging Lucia G, version 4.81). With the latter approx. 1000 objects were counted.

4. Results

4.1. Particle size and shape of oblate particles (example: pyrophyllite)

Fig. 1 shows the particle size distribution of pyrophyllite as measured via sedimentation (left curve, thin dotted) and via laser diffraction (right curve, thick full).

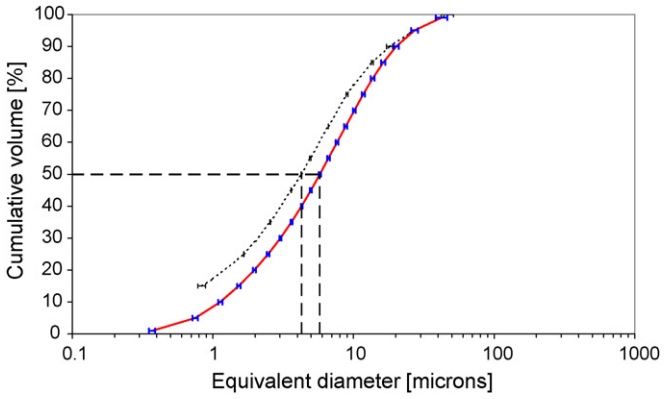

Fig. 1. Particle size distribution of pyrophyllite as measured via sedimentation (dotted curve, median 4.27 μm) and laser diffraction (full curve, median 5.76 μm).

It is evident that the laser diffraction values are larger than the sedimentation results, which allows an LS shape factor to be determined, cf. Fig. 2. The widely differing error bars in this SSD curve (shape-size dependence curve, Fig. 2) are a consequence of the increased size-determination uncertainty in the fine- and coarse-size region of both sedimentation and laser diffraction results (cf. the horizontal error bars in Fig. 1). In the fine-size region <1 μm the sedimentation results are subjected to error because of Brownian motion and the laser diffraction due to the breakdown of the Fraunhofer approximation, while in the large-size region (>25 μm for this sample) the curves of sedimentation and laser diffraction exhibit a cross-over, which clearly results in artefacts for the LS shape factor.

In the regions <1 μm and >10 μm (for this sample) the size-uncertainty of sedimentation and laser diffraction results is >5% (while the average is approx. 3%), which results in LS shape factors (empty squares in Fig. 2) with errors between 10 and 50%. The most reliable LS shape factors are between 4.0 and 5.3.

4.2. Particle size and shape of prolate particles (example: mesalamine)

Fig. 3 shows the SSD curve for mesalamine as determined via microscopic image analysis. The average aspect ratios of the

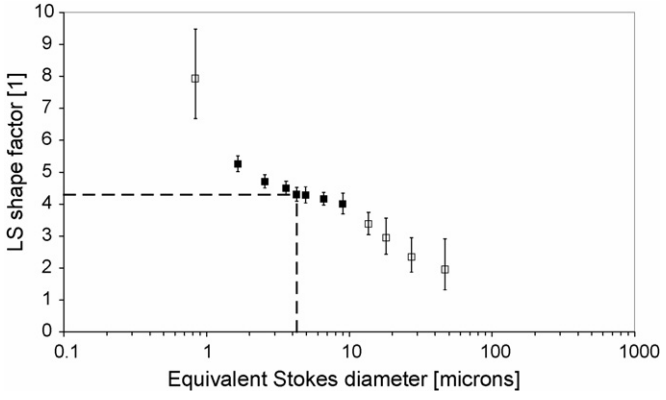

Fig. 2. SSD curve of pyrophyllite (median LS shape factor 4.3); full squares denote reliable values, empty squares probable artefacts.

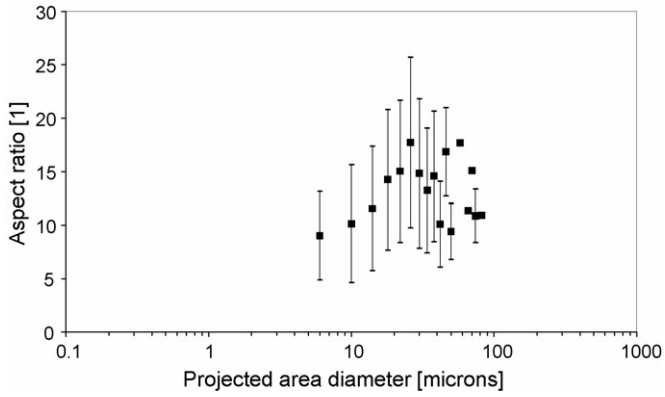

Fig. 3. SSD curve of mesalamine (size class average aspect ratios between 9 and 18, arithmetic average of individual particles approx. 12.5, grand arithmetic average of size class averages approx. 13.1 ± 2.8, median aspect ratio 8.9 ± 0.5).

individual size classes vary approximately between 9 and 18. Surprisingly, even such a large scatter in shape has a negligible effect on the results of the q_0–Q_3-transformation. The relative error is <2.5% in vertical direction, i.e. lower than the expected reproducibility of MIA measurements. Fig. 4 compares the size distribution measured by laser diffraction (average uncertainty approx. 8%) with the size distributions determined via MIA (after performing the q_0–Q_3-transformation). The size distribution measured by laser diffraction is broader than that measured via MIA and shifted towards smaller sizes, as expected, cf.[10] That means, although laser diffraction results can be used for a mutual comparison among different systems of prolate particles, it will not agree with MIA results (not even in the case of ideal cylinder or spheroid systems) and will usually show a median value lower than the MIA projected area diameter (here 16.9 μm versus 21.0 μm). Note, however, that the comparison between laser diffraction and MIA results performed in this contribution requires a q_0–Q_3-transformation[10] and is not allowed for the number-weighted primary results from MIA (although this is sometimes done in the literature).

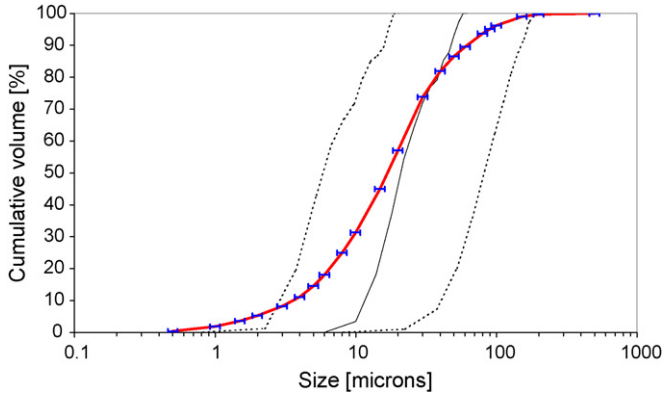

Fig. 4. Volume-weighted size distributions of mesalamine; dotted curves: minimum (left) and maximum (right) Feret diameter (MIA), thin full curve: projected area diameter (MIA), thick full curve with error bars: LD equivalent diameter.

5. Summary and conclusions

The size and shape characterization of oblate and prolate particles has been discussed with two typical examples, pyrophyllite (a ceramic raw material) and mesalamine (a pharmaceutical agent), which are representative for oblate (disc-like) and prolate (needle-like) particle systems, respectively.

For the first material (pyrophyllite), a convenient shape measure, the LS shape factor (approximately corresponding to some average aspect ratio of the system), can be extracted from a comparison of sedimentation and laser diffraction results. As expected, it was found that the LS shape factor is not size-invariant, i.e. it exhibits a shape-size dependence, when the average LS shape factor is given for individual size classes. The most reliable LS shape factors are obtained for the central region of the size distribution curve (here values between 4.0 and 5.3, with a relative error of 5–8%), while LS shape factors extracted from the peripheral regions of the size distribution curves (fine-size and coarse-size region) must be discarded.

For the second material (mesalamine) the size (projected area diameter, minimal and maximal Feret diameter) and the shape (aspect ratio) distribution have been determined by microscopic image analysis (MIA). It has been shown that even a relatively large variability of the average aspect ratio (here form 9–18) has only a negligible effect on the transformation of the MIA results to volume-weighted distributions. A comparison of (transformed) MIA results and laser diffraction data has shown that the size distribution obtained for prolate particles via laser diffraction is usually broader than MIA results and shifted towards the fine-size region, with a median value smaller than that of the projected area diameter distribution determined by MIA (16.9 μm versus 21.0 μm).

As a by-result, this investigation indicates that the reproducibility of laser diffraction data is better for oblate particles (±3%) than for prolate particles (±8%).

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References

1. Allen, T., (*5th ed.*). *Particle size measurement*, *Vol 1*Chapman & Hall, London, 1997.
2. Bernhardt, C., *Particle size analysis: classification and sedimentation methods*. Chapman & Hall, London, 1994.
3. Xu, R., *Particle characterization: light scattering methods*. Kluwer Academic Publishers, Dordrecht, 2000.
4. Russ, J. C. and Dehoff, R. T., *Practical stereology* (*2nd ed.*). Kluwer Academic/Plenum Publishers, New York, 2000.
5. Pabst, W., Kuneš, K., Havrda, J. and Gregorová, E., A note on particle size analysis of kaolins and clays. *J. Eur. Ceram. Soc.*, 2000, **20**, 1429–1437.

6. Pabst, W., Kuneš, K., Gregorová, E. and Havrda, J., Extraction of shape information from particle size measurements. *Br. Ceram. Trans.*, 2001, **100**, 106–109.
7. Pabst, W., Mikač, J., Gregorová, E. and Havrda, J., An estimate of orientation effects on the results of size distribution measurements for oblate particles. *Ceram.–Silikáty*, 2002, **46**, 41–48.
8. Lehmann, M., Berthold, C., Pabst, W., Gregorová, E. and Nickel, K. G., Particle size and shape characterization of kaolins—comparison of settling methods and laser diffraction. *Key Eng. Mater.*, 2004, **264–268**, 1387–1389.
9. Berthold, C., Klein, R., Lühmann, J. and Nickel, K. G., Characterization of fibres and fibre collectives with common laser diffractometers. *Part. Part. Syst. Char.*, 2000, **17**, 113–116.
10. Pabst, W., Berthold, C. and Gregorová, E., Size and shape characterization of polydisperse short-fiber systems. *J. Eur. Ceram. Soc.*, 2006, **26**, 1121–1130.
11. Pabst, W., Gregorová, E. and Berthold, C., Particle shape and suspension rheology of short-fiber systems. *J. Eur. Ceram. Soc.*, 2006, **26**, 149–160.