Particle size determination at different concentrated Al2O3 and SiO2 nanopowder suspensions with the ultrasonic spectrometer DT 1200

G. Steinborn[1], S. Scheler[2], J. Lehmann[2]
[1]Federal Institute for Materials Research and Testing Berlin, [2]University Bayreuth

1. Introduction:

The characterisation of particles in the nanometer range plays a deciding role in different industrial fields as pharmacy, nutrition technology as well as material science. The determination of nanoparticular powder is a great challenge for the measuring methods. With the ultrasonic and the electro acoustic spectroscopy the particle distribution down to 10 nm as well as the zeta potential can be measured. Additionally, those measuring methods provide an opportunity to analyse particle sizes in high solids concentrations.
In a 6 interlaboratory comparison the accuracy and reliability of the particle size distribution measurement are carried out. For this determination specific stabile nanosuspensions were selected using the ultrasonic spectrometer DT 1200 (Dispersion Technology Inc.).

2. Presentation of the measurement methods ultrasonic spectroscopy procedure:

The ultrasonic spectrometer DT1200 allows the determination of zeta potential and the determination of the grain size distribution of particles down to the nanometer range dispersed in aqueous and organic solutions and diluted and highly concentrated (1 - 50Vol. % solids) suspensions. During the measuring procedure ultrasonic attenuation, sound velocity and the acoustic impedance are determined. By mathematical modelling of these measurement results the particle sizes can be calculated.
The oscillation of charged particles in the acoustic field causes an alternating electric field. This forms the basis for the electro-acoustic measurements and allows the determination of zeta potential in these suspensions. By the characterization of the suspension with the ultrasonic spectroscopy a better assessment of homogeneity and occurrence of agglomerates of the various powders in the suspension can be achieved. It enables an improvement of the quality control process of manufacturing.
The particle size distribution for the example of a Silica-Ludox aqueous suspension (standard for particle size analysis) has been calculated from the ultrasonic attenuation as a function of frequency is shown in Figure 1.

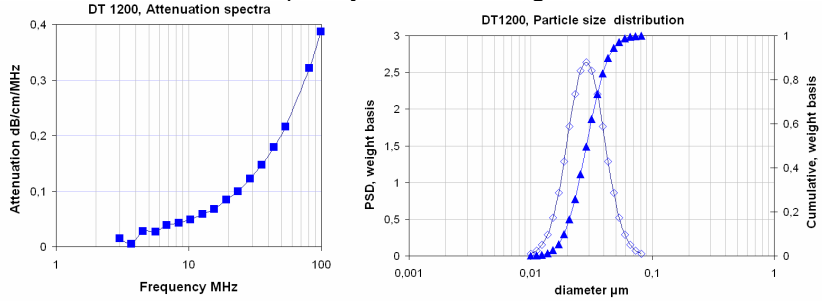


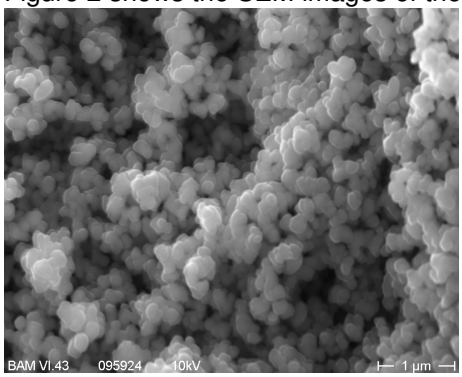
Fig. 1: Particle size analysis of aqueous 10 weight % Silica-Ludox suspension calculated from the measured attenuation spectra

The average grain size of the silica particles Ludox was determined at 29 nm for a monomodal particle distribution.

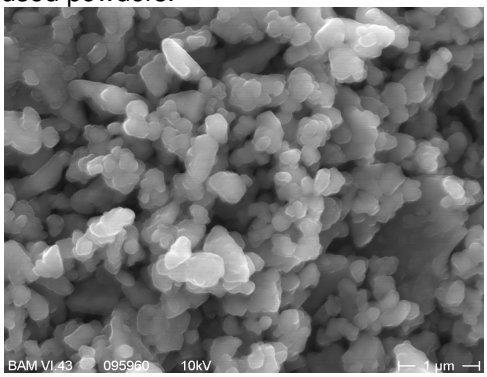
3. Determination of the particle size analysis

As tested objects, two commercial powders and two commercial suspensions were used (see Table 1) chosen as an example of common precursors in the ceramic and polymer chemistry. Additionally, the d_{50}-value of the samples is expected to be

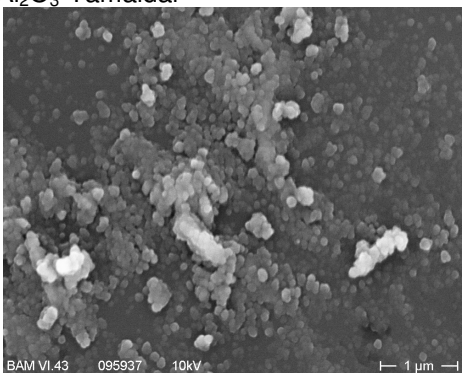
a unimodal particle size distribution in the nanometer and submicron particle range. Figure 2 shows the SEM images of the used powders.



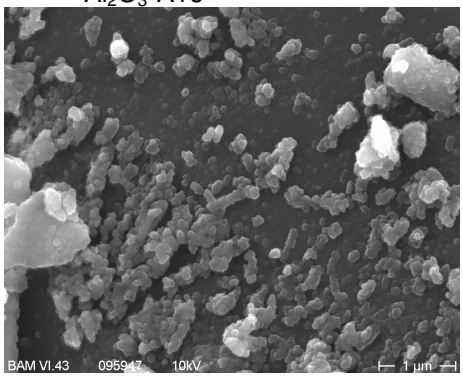
Al_2O_3-Tamaidar



Al_2O_3-A16



SiO_2-Koestrosol



SiO_2-Levasil

Fig. 2: SEM-images (scale 1µm)

Firstly, 2 vol. % Al_2O_3 suspensions (Al_2O_3-powder Tameidar; A16 alumina) were dispersed with the stabilizer CE64 (Zschimmer und Schwarz) in a 0.001 molar KCl-solution. The suspensions were treated for 30 minutes with the ultrasonic disintegrator Sonifier 450 of the company Branson (50% of maximum power, pulsed US50/50). Secondly, commercial available colloid dispersed SiO_2-suspensions (Koestrosol and Levasil) were measured with concentrations of 30 and 40 vol. %. Afterwards, the suspensions were determined in the ultrasonic spectrometer DT1200.

Table 1: Characteristics of the used samples

Sample	Production	d_{50}-value	Concentration vol.%
Al_2O_3-Tamaidar	Sumitomo; Japan	~150 nm	2
Al_2O_3-A16	Alcoa world alumina	~400 nm	2
SiO_2-Suspension (Koestrosol)	Bad Koestritz	< 100 nm	40
SiO_2-Suspension (Levasil)	H.C. Starck	< 100 nm	30

Previously, the optimal dispersion conditions of the Al_2O_3 powders were determined. For that, measurements of the Al_2O_3 suspensions were made in the DT1200 to determine the particle size at different additions of CE64 and dispersing times. For the Al_2O_3-Tameidar sample the addition of 1% CE64 and for the A16-alumina sample the addition of 0.6% CE64, based on the solids, showed the optimal dispersing in the aqueous suspension. Moreover, it was found that Al_2O_3 suspensions must be measured at the latest 2 days after manufacture. After the 2 days the suspension will sediment and agglomerate in spite of stirring and, thus, the experimental results will be changed with respect to the particle size. All

participants were given 6 randomly selected samples per suspension with a special marking.

4. Discussion of the results

For the cooperative test, no certified powders and suspensions were used as testing materials, so that the estimated parameters were determined as estimated values of random samples of all the cooperative test participants according to DIN ISO 5725-2, Item 4.2. There are no outliers using the Grubbs test. All measured values were used for the results.

Since the true values are not known, out of the cooperative test no ruling of the accuracy of the estimated values as well as of the individual laboratory results can be made. In contrast, the cooperative test leads to concrete results of the repeatability and the reproducibility standard deviation of the standardized measurement method.

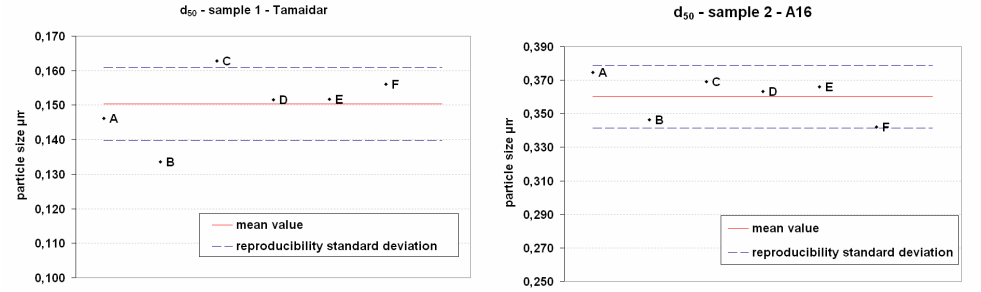
Using the formulas of the section 7.4 of DIN ISO 5725-2 the overall mean value and the precision value of repeatability standard deviation s_r and reproducibility standard deviation s_R were calculated (d_{50}-value for each of the 4 samples). The repeatability standard deviation is a dimension of the repeatability of the measurement results within the laboratories; determined as a weighted arithmetic mean of all laboratories. The reproducibility standard deviation is the estimated value of the scattering of the whole procedure and includes the deviation between the laboratories.

Both standard deviations are illustrated in the Table 2 below, supplemented by their relative amounts.

Table 2: Overview of the determined mean and precision values of the cooperative test (particle size distribution)

	sample 1- Tamaidar	**sample 2 – A16**	**sample 3- Koestrosol**	**sample 4 - Levasil**
	d_{50}	**D_{50}**	**d_{50}**	**D_{50}**
Mean value [µm]	0,1503	0,3602	0,0411	0,0338
repeatability standard deviation s_r [µm]	0,00405	0,01488	0,00222	0,00082
rel. s_r [%]	2,69	4,13	5,40	2,44
reproducibility standard deviation s_R [µm]	0,01055	0,01880	0,00289	0,00212
rel. s_R [%]	7,02	5,22	7,02	6,26

In Figure 4 the results of the individual laboratories (A to F) are graphically summarized.



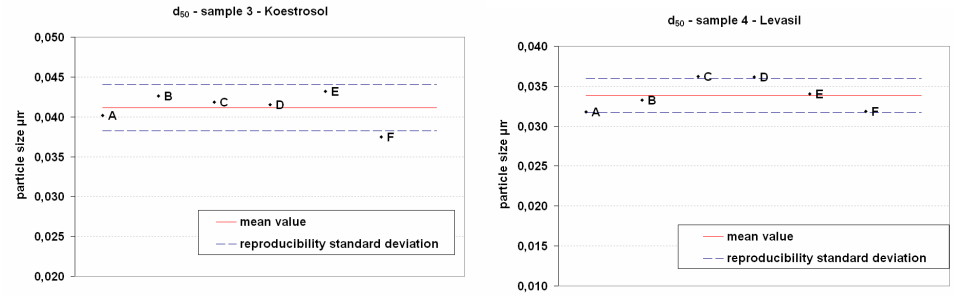


Fig. 4: d_{50}-value of the 4 determined samples

In sample 1, the calculated precision data show a good match of the d_{50} value of the repeatability standard deviation s_r with 2.69% and of the reproducibility standard deviation with 7.02% within the laboratories.

As well the sample 2 demonstrates good agreements for the d_{50} value of the repeatability standard deviation s_r (4.13%) and with 5.22% a good reproducibility standard deviation of the results.

The sample 3 (SiO_2 sol-sample Koestrosol) shows also good comparisons within the laboratories of the d_{50} value of the repeatability standard deviation s_r with 5.40% and of the reproducibility standard deviation s_R (7.02%).

Just as well, a good agreement could be seen for the d_{50} value of the repeatability standard deviation s_r (2.44%) and for the reproducibility standard deviation s_R (6.26%) in sample 4 (SiO_2 sol-sample Levasil).

5. Conclusion

Within the laboratories, the determined values of the repeatability standard deviation s_r are for the d_{50} value of sample 1 to 4 in the range of 2.44% (Levasil) to 5.4% (Koestrosol). The calculated reproducibility standard deviations show with the d_{50} value of particle size of ~ 7% a good match.

The evaluation of the cooperative test of the 4 samples showed in principle the quality of the measurement process and also the comparable results of different ultrasonic spectrometers (DT1200) for the determination of particle size distributions in aqueous suspensions with different solids concentrations in the range of nanometer or submicrometer.

6. Acknowledgments

We thank the staff of the laboratories (IPAT-Institute for Particle Technology Technical University Braunschweig, Technical University Dresden, Leibnitz Institute of Polymer Research Dresden, Department of Ceramic Materials Engineering University Bayreuth, Quantachrome) for participation in a cooperative test to determine the particle size of different suspensions.