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Grain size analysis with the slit ultramicroscope.

by J. Clif.

Translated from Staub 20:6 (1 Jun 60).

The search for an effective means to eliminate atmospheric dust from various dust-producing work processes includes the demand for highly accurate devices for the measurement of grain size and concentration of dust particles. Among a large number of methods (1), those permitting direct measurement in the suspended state without prior separation proved to be most feasible. These include several optical methods:
1. Registration of scattered light impulses emitted by dust particles during passage through a light beam which can then be classified electronically into grain classes. The instruments are complicated and bulky (2). 2. Grain size analysis by photometric evaluation of extinction measurements. This method fails when the dust is not monodisperse (3).
3. Visual observation under the microscope. Due to the small size of particles, darkfield illumination is required to contrast the light dust particles against a dark background, on which they may then be counted. Since dust particles move under the microscope, the field must be fixed photographically and the count is made later. Dirnagel (4) used this method to determine the rate of drop of dust particles within the grain range 0.3 - 100 microns. Richardson and Wooding (5) observe their material in a sedimentation chamber vertically to the line of drop, in which individual grain fractions appear in a stratified arrangement. Results of measurements confirmed the theoretical grain distribution in a particle system produced by gravity and Brownian movement (6). The range of grain sizes observed was 0.2 - 4 microns. In order to improve perception of very small particles, Saunders (7) proposed their enlargement through adiabatic expansion in the dust chamber and resultant adsorption of humidity. This produces new difficulties due to coagulation and the need of measurement of the adsorbed liquid. Zebel (8) describes a procedure for very small particles in the range near 0.1 micron with which he obtained close agreement with measurements of thermal precipitation.

Grain size analysis of mine dust involves the investigation of a relatively wide range of particle sizes and of mixtures of materials with widely dissimilar weights and optical properties. It did not seem feasible, therefore, to examine rates of drop, since the rates deviated from each other by several decimal powers. Sedimentation was therefore used for separation according to grain size; observations were made in the direction of sedimentation. In this connection I want to stress the fundamental difficulty of sedimentation analysis (3).

The difficulties inherent in the counting of dust particles on a photographic plate are compounded by the circumstance that the operator sees not only the brightly illuminated dust particles on the level adjusted by the objective lens, but also less distinct particles appearing as blurred discs on the margins of the illuminated zone. The delineation of particles that are to be counted represents a prime difficulty in their evaluation. The continuous transition between particles in focus and out of focus permits only subjective evaluation by the observer, resulting in erroneous counts. Another difficulty is presented by variable light intensities among larger and smaller particles. Different methods used in the development of exposed material will either cause particles of a certain brightness and magnitude to be still visible or no longer visible. This means that the lower limit of analyzable grain size is influenced by the technique used in the development of photographic material, and that an additional source of error is introduced thereby. Basically we can expect that grain sizes down to 0.2 microns in diameter will still be demonstrable by the darkfield method. A rough computation shows that illumination with an arc lamp scattered according to Rayleigh's model produces a light intensity which adequately blackens highly sensitive film. Computation according to Rayleigh's model seems justified, since the scattered light values obtained theoretically and experimentally by Senftleben and Benedict (9) in connection with coal particles do not deviate appreciably from those expected of Rayleigh's method in the angular range around 90° (cf. also Wolfsohn's contribution, 10). The calculation has been carried out according to Pohl's (11) instructions. The dielectric constant has been equated with 4 after Migashita (12) (cf. data on the index of refraction in 16). The starting values are set at an electrical power input of 750 W and 1% electro-optical efficiency. The illuminated zone has a diameter of about 0.5 x 6 mm vertically to the direction of light. The microscope's objective has an aperture A = 0.18 at 6-fold magnification. Total magnification was 60-fold. Thus dust particles with radius r = 0.1 micron produce a light intensity of about $2 \cdot 10^{-2}$ Lux on the exposed material, although light lost due to reflection from glass surfaces or due to filters and limitations in the field of view can only be estimated.

The sensitivity of the photographic material (Kodak Trix film) is listed as 25/10° DIN (German industrial standard). Under the present test conditions and method of development it may be assumed to be 30/10° DIN. Experimental variation of film brands and development technique failed to produce higher sensitivity in the film. At a minimal exposure time of 0.2 s, v. Angerer (13) obtains a minimal light intensity of $1.4 \cdot 10^{-3}$ Lux with which to exceed the threshold value. Thus, in the first approximation, the grain size of 0.2 microns in diameter may be considered the smallest grain size demonstrable with visible light. Using an interferometer, v. Baeyer and Gerhardt (14) reach the same conclusion; they also list 0.2 microns and claim to have increased sensitivity to 0.053 microns with the aid of ultraviolet light. Siedentopf and Zsigmondy (15) compute 6.4 millimicrons as the smallest visible particle diameter on the premise that the specific intensity of rays diffracted by the particle equals that of sunlight, i.e., about 10^9 HK/m^2. However, the authors themselves

deem the achievement of such intensities of scattered light impossible. For comparison, the present tests produced a scattered light intensity of about $2.5 \cdot 10^{-11}$ Watts per solid angle for a particle with radius 0.1 micron in the direction of sight. Assuming a surface of about 10^{-14} m^2, this corresponds to a luminous density of $5 \cdot 10^5$ HK/m^2. Moreover, the eye's limit of sensitivity must be considered far above that of the most sensitive photographic emulsions.

In characterizing the problems encountered in the photography of dust particles, it is to be noted that the intensity of scattered light in the range of Rayleigh's dispersion increases with the sixth power of the particle radius. The margins of exposure of a photographic emulsion, i.e., the interval between the threshold of veiling and maximal density, are only 1:10^4 (13), which means that in the presence of particles of widely disparate size the minimal exposure time required for small particles leads to marked over-exposure in the case of large particles.

Coordination of grain sizes and sedimentation rates was carried out with Stokes' law in its original form without correction terms (3):

$$U = \frac{2}{9} \frac{r^2 (\varrho K - \varrho L) g}{\eta}$$

Inclusion of Oseen's correction seems unnecessary, since the range of grain sizes perceived experimentally contained principally particles less than 5 microns in diameter. Measurements of larger particles made with the present instrument reveal relatively great instability for technical reasons, since the dust has not attained complete steadiness in the short period of 5 s between agitation and measurement. The employment of Oseen's correction term consequently does not seem indicated for this range. The table reproduced below lists radii of particles which during sedimentation have traversed at least the vertical distance h after time t. This means that only particles with a smaller radius than listed in Table 1 are present in the sedimentation chamber above level h at time t.

Table 1. Particle radius in microns at drop height h in cm and drop time t, $\varrho = 1.3$ g/cm^3

Drop height h (cm)		0.25	0.39	0.6	0.93	1.29	1.64	1.78
	5 s	1.79	2.24	2.78	3.44	4.06	4.60	4.78
	15 s	1.03	1.3	1.60	2.00	2.34	2.64	2.76
	30 s	0.73	0.91	1.13	1.41	1.66	1.87	1.96
	1 min	0.52	0.65	0.80	0.96	1.17	1.32	1.38
Drop time	2 min	0.36	0.46	0.57	0.70	0.83	0.94	0.97
	3 min	0.30	0.37	0.46	0.58	0.68	0.76	0.80
	5 min	0.23	0.29	0.36	0.45	0.52	0.59	0.62
	10 min	0.16	0.20	0.25	0.31	0.37	0.42	0.44
	15 min	0.13	0.17	0.21	0.26	0.30	0.33	0.36
	30 min	0.09	0.12	0.15	0.18	0.21	0.24	0.25

The instrument (Fig. 1) consists essentially of the Leitz slit ultramicroscope to which a semi-cylindrical sedimentation chamber with a height of 25 mm and a diameter of about 15 mm is attached underneath. A horizontal illuminating arrangement projects a narrow horizontal beam into the chamber. Microscopic observation is directed vertically from above, in the direction of sedimentation, during which particles falling through the illuminated zone assume a light contrast against the dark background. The path of illumination and observation contains facilities for the polarization of light. The sedimentation chamber is closed off in the direction of light entry and observation by thin, plane-parallel glass plates and is equipped for aspiration of dust-laden air. The photographic apparatus consists of a camera which permits supplemental, visual observation of the field (mirror reflex camera). Since the movement of dust particles in the field requires high shutter speeds, only highly sensitive films (Kodak Tri X, Agfa Isopan Record) are suitable for exposure. The light source consists of a lamp of high luminous density, e.g., arc lamp, searchlight, zircon oxide lamp, or an electronic flash. The test substance constitutes purest coal dust of known grain composition. Fig. 2 offers a total view of the apparatus.

The count is generally taken directly from the photographic negative, projected on a screen, if necessary. Various enlargements are made for control purposes. Fig. 3 shows an example of 10-fold linear enlargement of a photographic reproduction of the field.

Fig. 4 shows the compilation of a large number of test series. Listed is the number of particles which are smaller than a marginal grain size obtained during sedimentation. The graph shows, especially in the case of smaller grain sizes, that a considerable instability in measurements results from statistical fluctuations in particle count and possible errors in evaluation. A statistical evaluation by Neubauer of 19 independent measurements is shown by Table 2. The measured values are differentiated graphically, permitting separation into grain classes with identical class width.

Table 2

Class μ	$\bar{x}$ absolute	s_g absolute	$\bar{x}$ % of 150 (!)	s_g % of 150 (!)	s_{th} %	s_{exp} %
0 -0.5	15.2	8.36	10.1	5.55	2.46	4.96
0.5-1.0	20.4	8.71	13.6	5.80	2.80	5.08
1.0-1.5	19.7	5.52	13.1	3.68	2.75	2.44
1.5-2.0	17.1	4.00	11.4	2.66	2.59	0.61
2.0-2.5	15.4	4.41	10.3	2.94	2.48	1.58
2.5-3.0	13.6	3.77	9.1	2.52	2.35	0.91
3.0-3.5	11.2	4.10	7.5	2.73	2.15	1.68
3.5-4.0	8.5	4.58	5.7	3.05	1.89	2.39
4.0-5.0	11.7	4.84	7.8	3.22	2.19	2.36
5.0-6.0	8.1	2.57	5.4	1.71	1.85	0
6.0-7.0	5.4	2.34	3.6	1.55	1.52	0.30
7.0-8.0	2.5	1.87	1.7	1.25	1.06	0.66
	148.8		99.3			

Legend:

χ = median value,

s_g = total standard deviation,

s_{th} = theoretically expected standard deviation per equation

$$s_{th} = \sqrt{\frac{\chi_{\%} (100 - \chi_{\%})}{N}}$$

based on statistical principles,

N = total number of particles counted during one set of measurements (here: 150),

s_{exp} = standard deviation based on test procedure and evaluation according to the equation

$$s_{exp} = \sqrt{s_g^2 - s_{th}^2}$$

It is clearly evident, especially in the range of small grain sizes, that s_{exp} adds considerably to the total standard deviation, a circumstance attributable to the difficulties of evaluation already described. In the range of larger particles, on the other hand, total standard deviation is due essentially to statistical fluctuations; the latter could be reduced if the number N of counted dust particles were increased, but this would in turn compound the difficulties of evaluation. Moreover, it is evident that only a determination of median values from a relatively large number of independent measurements would lead to tolerably reliable information about grain distribution.

A translation of measurements into grain distribution according to weight percentages gave no agreement with an Andreasen analysis made of the same dust (Fig. 5). Andreasen's analysis yielded a much finer grain distribution. The explanation therefor might be found in the fact that dust is completely dispersed in Andreasen's sedimentation fluid. Blowing of dust into the ultramicroscope's dust chamber does not guarantee complete separation of all aggregates.

The following points are listed in conclusion:

1. Measurements with the slit ultramicroscope may be carried out directly at the site of dust inception, without prior sampling.

2. Individual measurements take relatively little time.

3. The achievement of sufficient precision requires evaluation of a large number of measurements.

4. Measurements can be made only in conjunction with photographic evaluation.

5. Photographic evaluation introduces new sources of error and involves relatively large expenditures of time.