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PREPARATION AND CHARACTERIZATION OF SOME PARTICULATE MATERIALS IN THE ALUMINUM INDUSTRY

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Abstract

Preparation and characterization techniques for the following particulate materials are reviewed: micromineralogical samples of bauxite, alumina and its trihydroxide, as well as sedimentary and respirable particles. Scanning electron microscopy (SEM), energy dispersive X-ray microanalysis (EDS) and image analysis were used to characterize the microminerals of bauxite. Comparisons were made among micromineralogical samples with various grain size fractions and the degree of weathering could be quantified. Programs were developed for characterization of the sandy and floury types of aluminum trihydroxide and alumina. We have used backscattered electron SEM images to characterize the size and shape of various aluminum trihydroxides. Comparison could be made between sandy and floury types of aluminum trihydroxide/alumina samples and the effects of technological changes could be quantified. The shape and composition of respirable particles in alumina plants were studied from the point of view of environmental protection.

Key Words: Particulate materials, scanning electron microscopy, energy dispersive X-ray microanalysis, image analysis, alumina, aluminum trihydroxide, bauxite, bauxite minerals, sedimentary particles, respirable dust.

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Introduction

Among the treated bauxite samples, the micromineralogical specimens enriched in detrital mineral species are of primary importance because their properties can give information on the bauxite genetics. These species are ilmenite, zircon, rutile, magnetite, etc., and their relevant properties are the total quantity, phase composition, size and shape. By studying these we can obtain information on the mother rock and the conditions under which the ore formed (Bárdossy, 1982). Since 1952 optical microscopy has been the common method in Hungary (Kiss, 1952) to identify the microminerals of bauxite, while electron probe microanalysis (EPMA) of bauxite dates back to 1973 (Bárdossy and Panto, 1973). Small and/or opaque minerals could not be unambiguously identified by optical microscopy. By means of EPMA mineral species smaller than 60 μm could be identified and the following mineral species were found: galenite, sphalerite and chromite. Nevertheless optical microscopy has remained the method most frequently applied in this field.

For past few years SEM is proving to be a powerful tool for the visual analysis of particulate materials in the aluminum industry. Since the seventies the morphological features of aluminum trihydroxide (Misra and White, 1971), its crystal growth and nucleation (Brown, 1972a), the role of secondary nucleation (Brown, 1972b) and the growth flaws (Gnyra *et al.*, 1974) have been studied by SEM. In order to meet the increasing demand of the modern electronic, ceramic and alumina industries it was necessary to develop methods for quantifying the size and shape of these particulate materials. On one hand, the modern electronic and ceramic industries need high purity and fine grained aluminum, while on the other hand, the modern aluminum industry requires high strength coarse grained aluminum.

The literature on preparation and characterization of samples of airborne particles for environmental studies is diverse (see e.g., Morgan and Winters, 1987; Grasserbauer, 1984). It appears that only the quantity of solid pollutants is monitored regularly because we

could not find any publication referring to the analysis of the composition of airborne pollutants in alumina plants. When presently new alumina plants and smelters are set up, careful environmental studies are carried out to choose and control the stages of production technology in order to limit the quantity of solid pollutants to an acceptable level (Nunes *et al.*, 1988).

The aim of this paper is twofold:

- to briefly summarize the literature of specimen preparation and characterization techniques relevant to the following 3 types of particulate materials:
  - micromineralogical specimens of bauxite,
  - alumina and its trihydroxide
  - sedimentary and respirable particulates of industrial pollutants;
- to present methods developed by the present authors together with some examples of applications.

Specimen preparation

Micromineralogical samples of bauxite

The preparation technique of micromineralogical samples of bauxite has been developed as a conventional silting method known in sedimentology (Kiss, 1952). The principle of this method is to convert the major iron containing minerals of the ore to ferrochlorides by treatment with hot hydrochloric acid and to remove them gradually by washing with distilled water and decanting the solution. The quantity of bauxite necessary for this treatment varies from about 100 g to 1 kg depending on the total quantity of detrital minerals. Generally the following grain size fractions of microminerals are obtained for optical microscopical studies by sieving: 0.32 - 0.2 mm, 0.2 - 0.1 mm and 0.1 - 0.06 mm.

Micromineralogical samples of Iszkaszentgyörgy (Hungary) bauxite for SEM and EDS studies and for classification were prepared by using several hundred gram material taken from various depths of three boreholes in the Iszkaszentgyörgy-Kincsesbánya Bito deposit (Csordás-Tóth *et al.*, 1985). The particulate samples were mounted on standard Cu stubs by conductive carbon paint and covered by a thin vacuum evaporated carbon layer (10-15 nm) for SEM and EDS studies. To obtain quantitative data on the size and shape of the particles and to classify the microminerals by chemical composition, the mineral grains with various size fractions were embedded into a two-component epoxy resin, and ground by SiC papers of 220, 400 and 800 mesh, respectively. Then they were polished with 0.3 μm alumina by machine and finally polished by hand on microcloth with aqueous MgO.

Alumina and its trihydroxide

Specimen preparation techniques including the application of various holder materials and dispersing agents are summarized in Table 1. These methods have been developed mostly for the purpose of characterizing the size and shape of alumina grains.

For SEM studies of aluminum hydroxide and alumina samples, either a small amount of powder or a preparation from an alcoholic suspension was poured on to the standard stubs covered by conductive carbon paint, and then coated by a thin Au or Au-Pd layer. Image analysis of backscattered electron (BE) images was done on embedded, ground and polished sections prepared by the procedure described above.

Sedimentary and respirable sample fractions

The principles and methods for immission type sample collection and analysis have been detailed in standards and in guidelines (ASTM D 1739-70, VDL 2119/4, VDI 2269/1). According to the ASTM standard D 1739-70, a simple open-topped cylinder with vertical sides and a minimum diameter of 13.5 cm, made of glass, stainless steel or plastic material, is appropriate to collect the settleable sample fraction. The vessel is half filled with water and the time period for collection is 1 month $\pm$ 2 days. Four vessels must be put up at each measuring place, the safety of the vessels and the free air flow should be ensured.

A simple and cheap procedure for the collection of sedimentary particulates is the "sticky" foil method (VDI-2119/4). A foil of 60x100 mm^2 area covered by vaseline is placed on a vertical rod of 1.5 m length. The collecting time is maximum 7 days, the detection limit is 0.4 mg/foil and 0.02 $gm^{-2}d^{-1}$ (the latter value refers to a 7 day collection period).

We have collected 22 sedimentary sample fractions by using collecting vessel mentioned above at various places of two Hungarian alumina plants and smelters. The places of sample collection were selected with the help of the leaders of the Environmental Protection Offices in the factories concerned. Fractions, soluble and insoluble in water, were collected together after they had settled down onto the surface of the collecting vessel during the unit time period, then filtered, decanted, dried and weighed carefully. Sedimentary particles were prepared from an alcoholic suspension on standard Cu stubs and were covered by a thin carbon layer for SEM and EDS studies.

The principles of microscopical studies have been laid down in VDI 2269/1. Various morphological features (including size and shape of individual particles and that of the agglomerates), optical, chemical and physical properties have been defined and summarized. Direct and indirect ways of sample preparation techniques have been distinguished by proposing methods best suitable for optical microscopic, SEM and transmis-

sion electron microscopy (TEM) studies. An Anderson Head cascade impactor designed for light microscopy was modified for SEM by Flickinger (1976). The application of glass fiber filters was not considered to be the optimum method for SEM and EDS studies because the surface of glass fiber filter is not smooth enough and also because the major constituents and even the trace elements in the filter can disturb the EDS analysis of the sample to be investigated. Particle removal by ultrasonic cleaning of the glass fiber has been proposed (McCrone and Delly, 1973).

Two methods using 0.1 μm Nucleopore and 0.1 μm Millipore type filters were studied by counting and sizing standard suspensions of short chrysotile fibers on TEM and SEM preparations (Millette *et al.*, 1978). Nucleopore membrane filters have also been proposed by Chatfield and Dillon (1978) for SEM and TEM studies. For the electron microscopic identification of small particles by EDS and electron diffraction techniques, Barbi and Skinner (1976) applied carbon coated "finder" grids which had the advantage that they improved the peak to background ratio for 2 μm particles.

Respirable particle fractions were collected mostly by an immission type high volume sampler designed by the Iron and Steel Research and Development Centre (Hungary, Budapest) for the collection of urban dust. This sampler has two impactor stages for separation of coarse particles and one membrane for the fraction finer than 5 μm. With regard to preparing and analyzing respirable particles the purpose of our work was to find materials and procedures best suitable for SEM and EDS studies. Experiments were performed by using polyamide fiber impactors and various membranes namely: 5-9 μm glass fiber, 5 μm teflon and mixed ester. The number, duration and pumping rate of sample collection were selected in such a way that dust of the mass to be evaluated could settle down both on the impactors and onto the membrane during an acceptable period (1/2-4 hours). The pumping rate varied between 30 and 65 m^3/h. Impactors and membrane together with the collected particles were stored in sealed polyethylene bags for weighing and for specimen preparation for SEM and EDS techniques. Direct and indirect methods were tried either by cutting small pieces from the membranes, sticking these onto standard Cu stubs and coating them by a conductive carbon layer, or by washing out the particles from impactors and the membrane by an ultrasonic treatment with alcohol, then drying the alcoholic suspension. Removing the particles by dry cleaning with a soft brush was also applied for polyamide fiber impactors with an efficiency of 30-60%. Particles settled down on the mixed ester membrane were removed by ultrasonic agitation with Calgon (Na-hexametaphosphate) diluted with distilled water to 0.05%.

Table 1: Preparation Methods for Particulate Alumina Samples.

Specimen holder/dispersing medium	Treatment	Reference
aluminized glass slides/diluted NH_4OH		White et al (1970)
glass slides covered by Au or Al/eutectic composition of camphor and naphthalene		Thaulow and White (1971/72)
/distilled water	ultrasonic agitation	Johnson, Jr. et al (1972)
polished Ge/eutectic composition of camphor and naphthalene		White et al (1974)
	embedding in Spurs medium, grinding, polishing	Dinger and White (1976)
	embedding into a two component epoxy resin, grinding, polishing	Csordás-Tóth et al (1986)

For the preparation of the particles removed from impactors and membranes two types of dispersing medium were tried: dibutyl amine (diluted with distilled water to 0.5 ml/l) + tensiofix + dimethylamine (also diluted to 0.5 ml/l with distilled water) and Calgon. The particle concentration in preparations placed as drops on the sample holder was kept in the range of 0.5-5 mg/ml. Particulate samples prepared in this way were covered by a vacuum-evaporated thin carbon layer.

Instruments and Methods

A Philips SEM-505 scanning electron microscope (SEM) was used both in secondary and backscattered electron mode at an accelerating voltage of 25 kV and a specimen current of 10^{-12} or 10^{-9} A. EDS analysis of micromineralogical samples of bauxites was carried out by an EDAX 711 energy dispersive X-ray spectrometer (with a 26° take-off angle) attached to the SEM, while sedimentary and respirable particles were analyzed by a Link AN 10/55 energy dispersive microanalyzer (with 40° take-off angle) at 20 kV. Characterization of alumina and its trihydroxide grains by their sizes and shapes was carried out by an Omnicon Reichert FAS-II image analyzer attached to our SEM by an EM-II module.

Classification of the bauxite minerals present in the micromineralogical samples was done by the image analyzer.

Method of classification of microminerals by their chemical composition

Optical microscopic and SEM and EDS studies of numerous individual grains of micromineralogical samples established a basis for the classification of the grains by their chemical composition. Simultaneously the mean values of some geometrical data (length, width, area and perimeter of the sectioned grains) and shape factors characterizing the degree of weathering were determined. Since the method is described in detail in Csordás-Tóth *et al.* (1985), only a short summary is given here. First the backscattered electron image was converted from analog to digital in such a way that the image frame was decomposed into 256 x 256 pixels, while the beam was staying 12 μsec at each image point. Then the following 12 geometrical parameters of the grains present in the image frame were measured: x and y coordinates of the centre of mass of the grains, area with and without holes, real and convex perimeters, length, width, Feret diameters at 0 and 90°, respectively, projected length and the maximum horizontal chord, respectively. Then the electron beam was directed back to the centre of mass of each grain and the net intensities of the following X-ray lines were determined: Al K_α, Si K_α, Zr L_α, K K_α, Ti K_α and Fe K_α, respectively. Spectrum acquisition was performed for a minimum of 10 seconds. After HCl treatment of the Iszkaszentgyörgy bauxite the remaining particulates were classified as follows: Al-rich, quartz, zircon, K-feldspar, Ti-rich, ilmenite, Fe-rich and miscellaneous. The basis for this classification is the following: "pure" chemical classes were called those in which one major element was present and the others were at the trace element level. In "mixed" classes the classification was done on the basis of the intensity ratio of two or three major elements, e.g.:

for zircon: the Zr L_α line has the maximum intensity and the intensity (I) ratio $I_{Zr}/I_{Si} > 1.5$;

for K-feldspar: the Si K_α line has the maximum intensity and $I_{Al}/I_{Si} > 0.1$; $I_K/I_{Si} > 0.1$;

for ilmenite: $0.4 < I_{Fe}/I_{Ti} < 2.5$.

Classification of the aggregates/agglomerates of aluminum trihydroxide and alumina by their size and shape

There is a large literature dealing with the quantification of the size and shape of particulates including aluminas. A contour analysis technique was developed by Matson *et al.* (1970) for the computer processing of SEM images and this put a basis for the further developments. White *et al.* 1972 developed a fully automatized

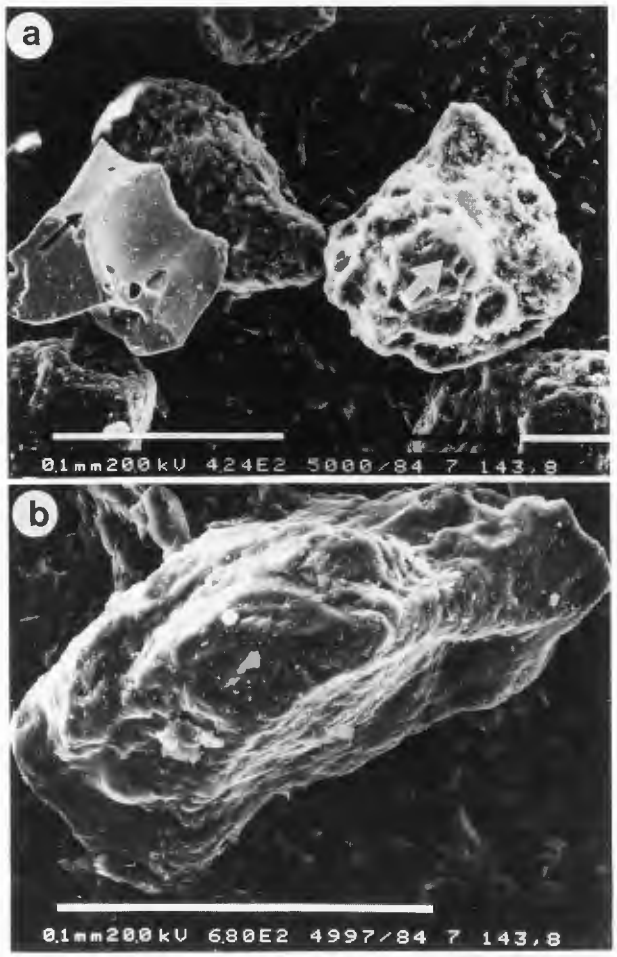

Figure 1a. Grain rich in Al remaining from the bauxite (class 1); fraction: 0.32-0.2 mm, depth: 143.8-144.8 m.

Figure 1b. Quartz (class 2); fraction: 0.32-0.2 mm, depth: 143.8-144.8 m.

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Figure 1c. Zircon (Zi, class 3); fraction: 0.1-0.06 mm, depth: 195.6-196.6 m.

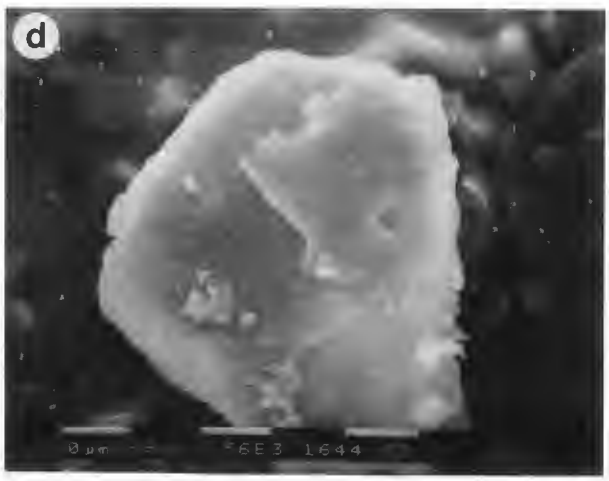
Figure 1d. Slightly eroded, broken feldspar (class 4); fraction: 0.1-0.06 mm, depth: 199.6-200.6 m.

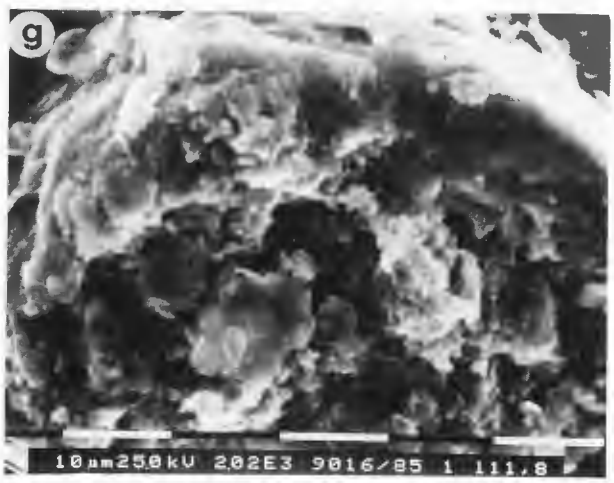
Figure 1e. Ti-rich (probably rutile; marked by arrow) grain (class 5); fraction: 0.1-0.06 mm. depth: 199.6-200.6 m.

Figure 1f. Ilmenite (class 6); fraction: 0.1-0.06 mm, depth: 192.6-193.6 m.

Figure 1g. Fe-rich grain, highly eroded (class 7); fraction: 0.32-0.2 mm, depth: 111.8-112.8 m.

Figure 1h. Ilmenite grain with apatite (Ap) and K-feldspar (Fp) inclusions.





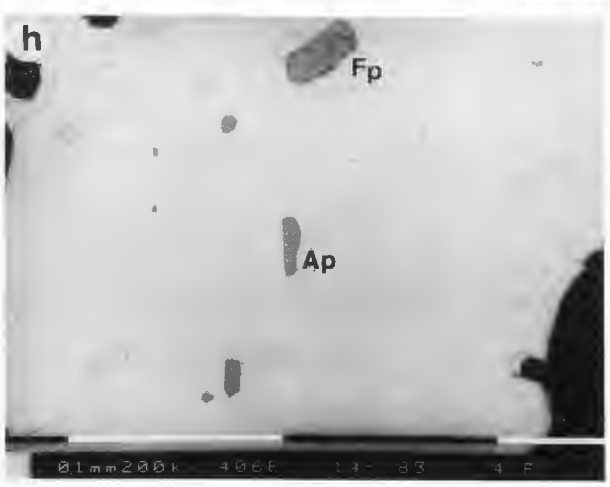
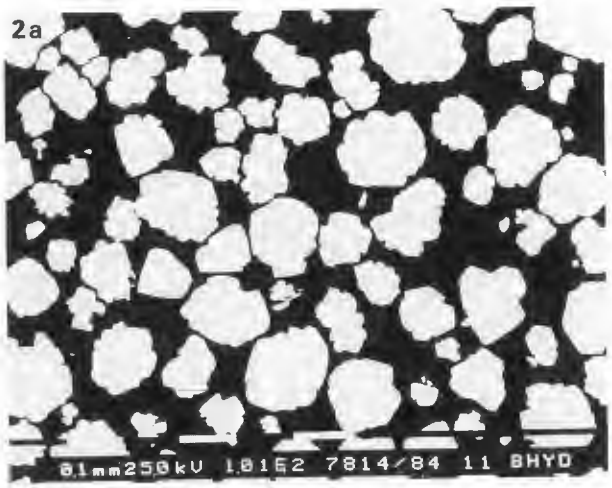
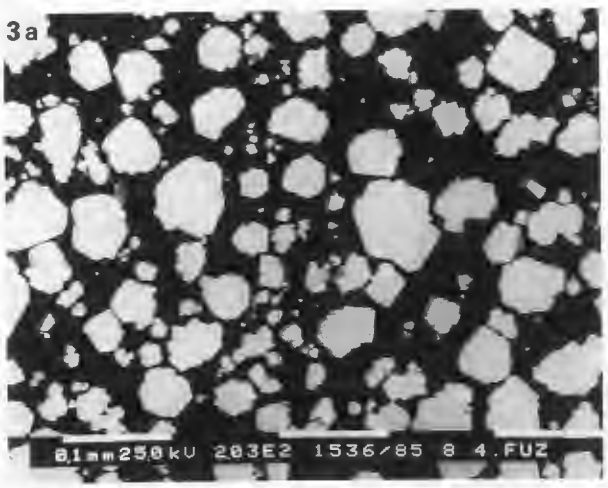
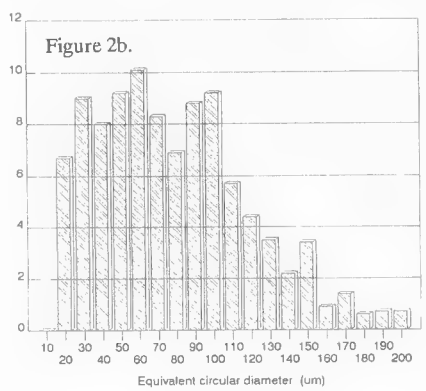


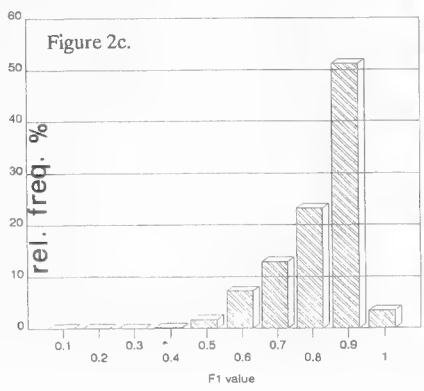
image analytical technique for the evaluation of the size and shape of relatively simple alumina grains. Secondary electron images of various alumina samples were evaluated and grain size distribution curves and histograms of different shape factors were obtained. Quantitative 3-D characterization of particulate materials including alumina grains and sintered alumina was performed by Dinger and White (1976) by using polished sections and various models optimized for the shape of the particles (e.g., sphere, cube, octahedron, etc.).

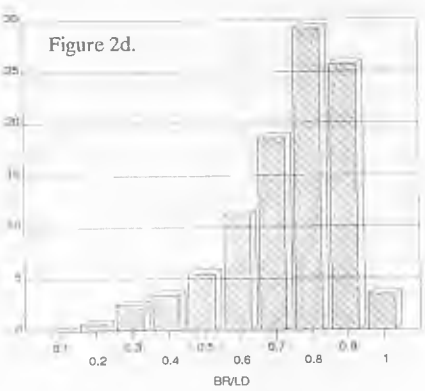
Two types of image analytical technique were developed and applied by Hsieh (1985) for technical grade aluminum trihydroxide grains. The shape factor of 4π area^2/perimeter was measured by a semiautomatic image

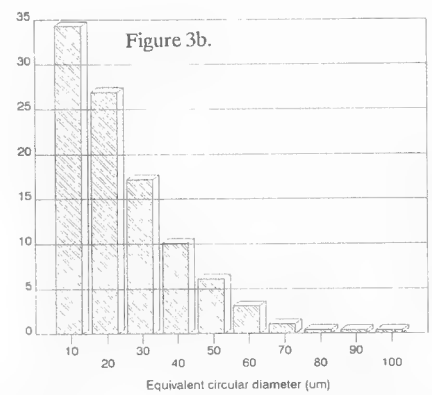


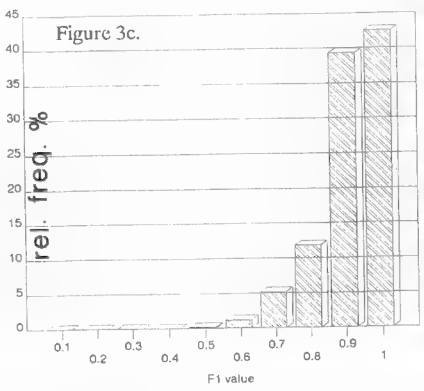












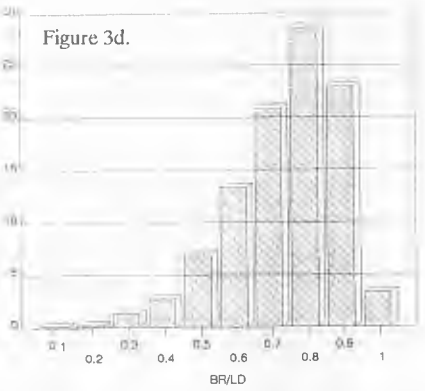


Figure 2: Backscattered electron image (2a) and histograms of the grains according to their equivalent circular diameter (2b) and shape factors of F1 = $2\sqrt{\pi}\sqrt{area/perimeter}$ (2c) and F5 = width/length (2d) for a sandy type of aluminum trihydroxide sample.

Figure 3.: Backscattered electron image (Fig. 3a) and histograms of the grains according to their equivalent circular diameter (Fig. 3b) and shape factors of F1 = $2\sqrt{\pi}\sqrt{area/perimeter}$ (Fig. 3c) and F5 = width/length (3d) for a floury type of aluminum trihydroxide sample.

analyzer on secondary electron images with a light pen. The other method was based on the Fourier analysis of the particle profile. Significant differences could be observed among the aluminum trihydroxide grains by means of this method.

We have written measuring and evaluating programs for the Reichert Omnicon FAS II, to characterize aggregates and agglomerates of aluminum trihydroxide and alumina samples. These programs could be run either on-line or off-line (Stefániay *et al.*, 1984).

Twelve geometrical data of each grain were measured by means of our program and all data were stored on a floppy disk for further evaluation. The measured parameters were as follows: area with and without holes (AF and AA, respectively), real and convex perimeters (PR and CP, respectively), longest dimension (LD), width (BR), projected length (PL), maximum horizontal chord (MC), x and y coordinates of centre of mass of the grains, and Feret diameters at 0 and 90 degrees, respectively. By our method about 1000 grains can be measured in an acceptable time (30 minutes to 1 hour). Shape factors are defined by Réti (1983) as dimensionless quantities smaller than or equal to 1 for characterizing the shape of the grains. By means of mathematical procedures many shape factors can be derived, for each type of material there are optimal shape factors. In case of aluminum trihydroxide and alumina of smelter grade the following shape factors were found to be important:

1. The shape factor calculated by the formula $F1 = 2\sqrt{\pi}\sqrt{area}/perimeter$ is used to characterize the degree of departure from a perfect circle. This shape factor has a value of one for a circle, 0.778 for an equilateral triangle, 0.886 for a square, 0.952 for a hexagon, 0.835 for a rectangle with a LD/BR ratio of 2 (Hsieh, 1985). The more complicated the perimeter of a feature, the smaller is this shape factor. Hsieh (1985) used the square of this shape factor for characterization.

2. The F4 = CP/PR ratio is a good measure of the grain shape complexity. In the case of a circle or simple convex figure, this shape factor has a value of one or very near to one, but in case of aggregates formed from a large number of prismatic crystals this shape factor differs from one.

3. The F5 = BR/LD formula gives information on the elongation of the grains. In case of elongated and/or irregular grains it has a smaller value than for compact agglomerates.

Different types of evaluation programs were written. One of them gives the relative frequencies by number and the average shape factors of aluminum trihydroxide/alumina grains for each selected equivalent circular diameter interval (the equivalent circular diameter is calculated from the measured projected area without holes). In addition, histograms of some shape factors can be obtained. Another program for evaluation can give mean values and deviations of eight important geometrical data (AA, AF, LD, BR, PL, PR, CP, MC) and of shape factors together with the result of classification by the equivalent circular diameter.

Results and Discussion

Microminerals of Iszkaszentgyörgy bauxite

Figure 1 shows secondary electron images of the typical grains (quartz, ilmenite, K-feldspar, Al-rich, Ti-rich, Fe-rich and small amounts of miscellaneous species) found in the micromineralogical samples of Iszkaszentgyörgy bauxites. Among them apatite (containing chlorine) and K-feldspar were detected as inclusions in ilmenite. The maximum size of apatite inclusions was about 30 μm. Apatite was more frequently detected in ilmenite grains than feldspar. These inclusions of ilmenite are proving the magmatic origin of an ilmenite variation, which is consequently a detritic (allotigenetic) micromineral of this bauxite. In addition to these microminerals calcium sulfate (probably gypsum), grains with Cu, Zn and Ni content and grains enriched in manganese, iron and calcium were also found in the samples of the three boreholes (Csordás-Tóth *et al.*, 1985).

Micromineralogical samples taken from the bauxite of borehole No. 18 were classified by their chemical composition. Quantitative data gained by our programs are summarized in Table 2. for sample fractions of 0.32-0.2 mm and 0.1-0.06 mm, respectively.

In the fraction of 0.32-0.2 mm size quartz was found to be the dominant constituent (70.3%) while in the smallest grain size fraction ilmenite was the most frequent micromineral (70.4%). As to the geometrical parameters of the grains ilmenites proved to be the largest ones. Ilmenite grains of this fraction were significantly eroded, relief of smooth hexagonal plates was hardly found. The morphology of the feldspar grains also showed the effect of strong weathering. Consequently the ratio of the convex and real perimeter was found to be the smallest for feldspar and largest for quartz.

In the smallest grain size fraction the grains enriched either in Ti or Fe showed the largest degree of weathering while zircon did not suffer from that phenomenon. Zircon was found not only in the form of individual mineral grains of the studied samples, but also intergrown with ilmenite crystals (A. Csordás-Tóth *et al.*, 1985). An oriented intergrowth of zircon and ilmenite has to be assumed during bauxite formation, i.e., crystallization under normal pressure-temperature conditions has to be assumed.

Table 2.

Average geometrical data of chemical classes of microminerals

Fraction: 0.32-0.2 mm

C	N (%)	AA (μm^2)	LD (μm)	BR (μm)	CP/PR	F1
1.	14.1	28520	265	137	0.89	0.74
2.	70.3	28670	233	142	0.92	0.75
3.	-					
4.	6.3	36933	350	160	0.81	0.65
5.	-					
6.	8.0	50600	375	197	0.88	0.83
7.	1.3	858x	47	23	0.96	0.74

Fraction: 0.1-0.06 mm

C	N (%)	AA (μm^2)	LD (μm)	BR (μm)	CP/PR	F1
1.	-					
2.	-					
3.	8.2	1523	65	32	0.94	0.7
4.	-					
5.	4.1	610	40	20	0.88	0.74
6.	70.4	1430	61	34	0.87	0.75
7.	17.4	1310	54	35	0.81	0.72

C: number of chemical classes;
N: percentage by number of particles;
AA: average area of particles without holes;
LD: average longest dimension of particles;
BR: average breadth of particles;
CP/PR: average ratio of convex and real perimeters of particles;
F1: average ratio of square root of the area and real perimeter of particles (Saltikov shape factor);
x: mixed by some percentages of grains with small grain size.

Table 3.

Granulometric properties of floury types of aluminium trihydroxide samples produced under different conditions

Mark of the sample	Production conditions	Result of sieving -45 μm (%)	Attrition index (%)
1.	agglomerated in the plant by batch process 72°C/6h, seed ratio: 0.2	38	32.2
2.	Sample №1. reagglomerated in laboratory 75°C/6h, seed ratio: 0.2	18	12.2
3.	seed aluminium trihydroxide 56°C/50h	54	13.0
4.	agglomerated in a liquor containing 6 g/l SO_3 75°C/6h, seed ratio: 0.2 56°C/48h, seed ratio: 3.5	-	-

Cont. of Table 3.

Mark of the sample	F1 range	Image analytical data Shape factors F1 Rel. frequency (%)	F5 range	F5 Rel. frequency (%)
1.	0.0-0.6	0.9	0.0-0.6	17.4
	0.6-0.7	4.2	0.6-0.7	20.0
	0.7-0.8	16.3	0.7-0.8	33.7
	0.8-0.9	59.8	0.8-0.9	26.3
	0.9-1.0	18.8	0.9-1.0	2.6
2.	0.0-0.6	2.7	0.0-0.6	26.5
	0.6-0.7	6.5	0.6-0.7	22.9
	0.7-0.8	16.2	0.7-0.8	29.4
	0.8-0.9	43.0	0.8-0.9	19.9
	0.9-1.0	31.6	0.9-1.0	1.2
3.	0.0-0.6	1.3	0.0-0.6	23.1
	0.6-0.7	5.1	0.6-0.7	18.6
	0.7-0.8	11.8	0.7-0.8	29.1
	0.8-0.9	39.3	0.8-0.9	25.7
	0.9-1.0	42.5	0.9-1.0	3.5
4.	0.0-0.6	8.7	0.0-0.6	38.5
	0.6-0.7	10.5	0.6-0.7	20.0
	0.7-0.8	20.5	0.7-0.8	26.5
	0.8-0.9	43.3	0.8-0.9	14.3
	0.9-1.0	17.0	0.9-1.0	0.7

Aluminum trihydroxide and alumina samples

Classification of the grains by their equivalent circular diameters and histograms of two shape factors are shown in Figures 2 and 3 for a typical sandy and floury type of aluminum trihydroxide sample. The most significant difference is obviously that of the size of aggregates/agglomerates present in the given samples. Also differences can be found in the histograms of the shape factor Fl characterizing the degree of departure from a perfect circle. If we consider the F1=0.8-1 range, then for floury aluminum trihydroxide 70% of the total number of the grains falls in this range, while for sandy aluminum trihydroxyde this value is only 54.6%. The explanation of this phenomenon is the agglomeration process which promotes the increase of large sized grains (Anjier and Marten, 1982). A slight difference can be found in the histograms of the shape factor characterizing the degree of elongation. This is in accordance with observations made by numerous authors that the grains of aluminum trihydroxide are only slightly elongated. In order to get significant differences in the shape of aluminum trihydroxide/alumina grains the method of Fourier analysis of the grain profile (Hsieh, 1985) should be applied. The method developed and applied by the authors is not able to get such big differences in the shape of individual grains but the tendencies and the effect of changes in production technology could be followed and quantified. It was tried to quantify the following phenomena and effects by our image analysis procedure (Csordás-Tóth *et al.*, 1986):

- agglomeration-re-agglomeration
- the effect of sulfate content and
- that of the organic impurities of the caustic liquor on the compactness of the grains in aluminum trihydroxide samples.

Here only the studies of agglomeration are dealt with due to the importance of this phenomenon for the production of sandy aluminum trihydroxide and alumina. In Table 3 some results of our image analysis method are shown together with the results of sieving and that of the attrition test for selected floury type of aluminum trihydroxide samples. On the basis of the measured attrition indices of sample No. 1 and 2 an enhancement of the strength of re-agglomerated grains was found. According to the image analysis data, this enhancement can not only be explained by the more compact form of all grains, but also by the decrease of the number of grains below 10 μm and between 10 and 20 μm (see Figure 2 in the conference material of Csordás-Tóth *et al.*, 1986) and by the fact that the bigger grains (in sample No. 2) are closer to the stable sphere form (in a section to a circle) than the grains of the original sample No. 1. This is illustrated by the following data of the image analysis: the relative frequency of grains having Fl =0.9-1 has increased from 18.9 to 31.6%.

Note: The definition of the attrition index (AI) is as follows:

$$AI = (X-Y)*100/X$$

where
X = % -45 μm before attrition test,
Y = % -45 μm after attrition test,

Sedimentary and respirable fractions of samples collected in Hungarian alumina plants and smelters

In one of the Hungarian alumina plants generally the emission source of corundum played an important role. Sharply edged corundum grains with elongated forms can be seen in Figures 4a and 4b, respectively, in the case of samples collected at the calcining furnace and even in the car parking. In addition, the effect of fine sodium aluminate came into force and quartz, fine SiO_2 could be found at refractory plants. Smelters could be characterized by the emission of calcite and dolomite (from soda-lime plants), fine aluminum trihydroxide and alumina. Grains with sharp edges in samples taken from the tool workshop might have a disadvantageous effect upon the surface finishing (see Figure 4c). Grains of flying ashes originated from a power plant near to the selected smelter could also be detected. One example is shown in Figure 4d. Small amounts of Fe, Ca and S were also found by EDS analysis in addition to Al and Si. The flying ash grain situated in the middle of the secondary electron image is probably mullite (found also by Mattigod, 1982).

Energy-dispersive analysis of the samples taken at the high pressure foundry, electrolysis bath and in the tool workshop always showed S on the surface of the grains, while Pb was found in the samples of the tool workshop and that of the high pressure foundry. By optical emission spectroscopy carried out on the samples of the electrolyzing bath and that of the high pressure foundry apart from Pb also Ni, Ti, Na, Zn, Cu were detected.

Secondary images of respirable particles from a refractory plant prepared by an indirect method using distilled water and Calgon dispersing agent are shown in Figure 5 for the case of an impactor and that of a teflon membrane. The size difference could, however, be easily recognized in these images. Transmission electron microscopy would be a more appropriate method to study the individual particles of respirable fractions. Diatomite grains with spherical and sometimes comb-like form could often be found in samples taken from the refractory plant (see Figure 6).

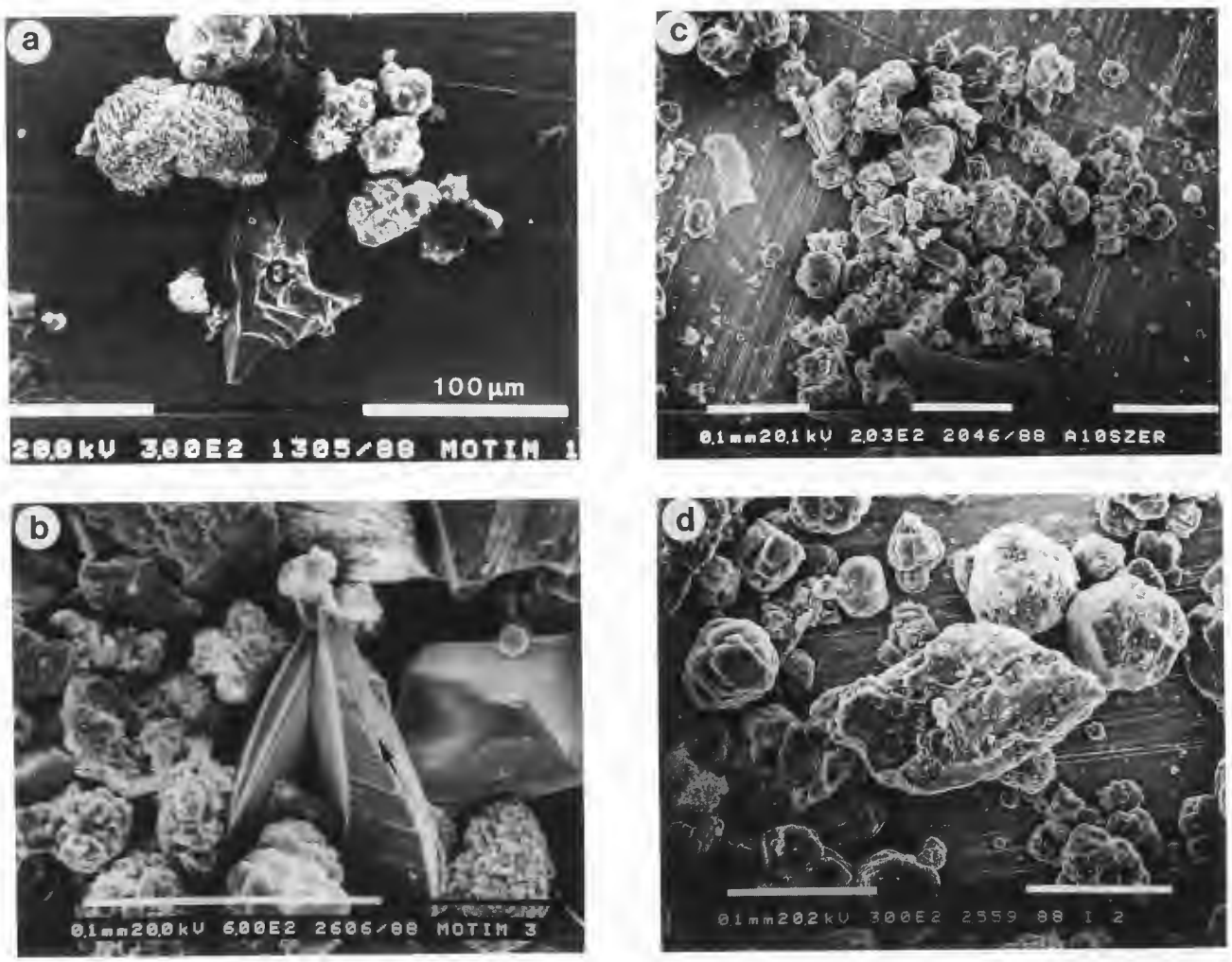


Figure 4: Secondary electron images of sedimentary particles collected at various places in Hungarian alumina plants and smelters.

Figure 4a: Grain marked by C electrocorundum with sharp edges. The sample was collected at the calcining furnace.

Figure 4b: Grains with sharp edges are also corundum. The sample was collected at a car parking of an Hungarian alumina plant.

Figure 4c: Secondary electron image of a sample collected in a tool workshop also shows grains with sharp edges dangerous for the surface finishing.

Figure 4d: EDS analysis of the sample shows the presence of flying ash originated from a power station near to the smelter. The sample was collected at a pot room of a Hungarian smelter.

Summary

Three types of particulate materials of importance in the alumina industry were studied by SEM, EDS and image analysis techniques - microminerals of bauxite - aluminum trihydroxide of smelter grade, - sedimentary and respirable fractions of samples collected in Hungarian alumina plants and smelters.

First we reviewed specimen preparation and classification techniques. The parameters of sample collection and those of the preparation techniques for SEM and EDS studies were determined for the respirable sample fractions by using a high volume immission type sampler. We have written computer programs for the classi-

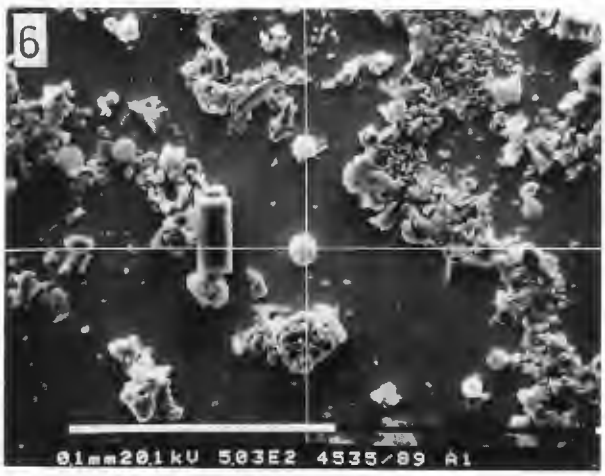


Figure 5: Secondary electron images of respirable particles collected by an immission type high volume sampler in a refractory plant in Hungary.
Figure 5a: Sample taken of one of the impactors.
Figure 5b: Sample taken from a teflon membrane.
Figure 6.: Secondary electron image of a sample containing diatomite grain marked by a cross.

fication of microminerals of bauxite based on their chemical composition and geometrical data. Results obtained by this type of classification can give information on the bauxite genetics in addition to that obtained by optical microscopy. Image analysis programs developed for the classification by the equivalent circular diameter and for the determination of shape factors can be useful to follow technological changes in the production of smelter grade aluminum trihydroxide/alumina samples. Results obtained by the combination of various analytical methods on the sedimentary and respirable sample fractions collected in Hungarian alumina plants and smelters have drawn the attention not only to the quantity but also to the quality of these particulate matters, i.e., the size, shape, and chemical composition.

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Discussion with Reviewers

H.P. Hsieh: Unless a special scheme is implemented in the image analysis program, two or more particles contacting or overlapping each other are taken as a large particle. Has precaution been taken to address the problem?

Authors: Unfortunately the software of our OMNICON system did not take into account the touching and overlapping particles. We are aware of this problem and we tried to counteract it by a suitable preparation technique. As you can see in Figures 2a and 3a, resp. this preparation technique gives a quite good dispersion of the grains.

M. Patel: As the temperature of seeding decreases one would expect increase in particle size but results of -45 μm of expt No. 1 and 2 carried out at 72 and 75 °C show it to be the reverse. How can this be explained?

Authors: Sample No.1 was made under industrial circumstances by a batch precipitation process, while sample No. 2 was produced in the laboratory. The increase in the crystal size can be explained partly by the agglomeration process, partly by a slight crystal growth which can be promoted by the increase of the temperature.