# ARCHIVESOFENVIRONMENTALPROTECTIONA R C H I W U MO C H R O N YŚ R O D O W I S K Avol. 32no. 1pp. 23 - 322006

PL ISSN 0324-8461

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# GRANULOMETRIC COMPOSITION OF DUST RELEASED FROM ZINC AND LEAD SMELTING

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Keywords: lead and zinc smelting; particle size, granulometric composition, statistical analysis.

# SKŁAD ZIARNOWY PYŁU POWSTAJĄCEGO W PROCESACH TECHNOLOGICZNYCH HUTNICTWA CYNKU I OŁOWIU

Przeprowadzono badania nad składem ziarnowym pyłu pochodzącego z głównych węzłów technologicznych jednej z hut cynku w Polsce, a mianowicie ze spiekalni, pieca szybowego i rafinerii ołowiu. Próbki pyłu pobrano z pyłu zatrzymanego w filtrach workowych. W badaniach zastosowano analizę sitową, sedymentacyjną i laserowy analizator składu frakcyjnego. Określono udział liczbowy i masowy pyłu występującego w dziewięciu przedziałach ziarnowych:  $0-2, 5, 2, 5-5, 5-10, 10-20, 20-32, 32-40, 40-63, 63-100, 100-250 \mum. Uzyskane wyniki poddano, stosując metodę estymacji nieliniowej, analizie statystycznej, której celem było wyznaczenie funkcji matematycznych opisujących zmiany składu frakcyjnego poszczególnych pyłów (liczbowego i masowego). Zastosowanie modelu regresji w postaci wielomianowej zapewniło wysoki stopień korelacji dopasowanych funkcji do danych pomiarowych. Wyniki badań przedstawiono w tabelach i w formie graficznej. Stwierdzono znaczne zróżnicowanie składu frakcyjnego badanych próbek pyłu zależnie od mechanizmu tworzenia się pyłu, a tym samym od warunków prowadzenia procesu technologicznego oraz składu przetwarzanych surowców. Duży udział cząstek respirabilnych w pyle może stanowić powaźne zagrożenie dla populacji, zwłaszcza w przypadku awarii urządzeń odpylających w instalacjach produkcyjnych.$ 

#### Summary

Granulometric composition of dust coming from main technological nodes – i.e. from the sinter plant, blast furnace and lead refinery – of the zinc smelter in Poland was investigated. Samples of dust trapped in bag filters were collected. The sieve analysis, sediment analysis and a laser analyzer of granulometric composition were used in investigations. Mass and particle number contributions of the dust to nine granulometric fractions having aerodynamic diameters in intervals: 0-2.5, 2.5-5, 5-10, 10-20, 20-32, 32-40, 40-63, 63-100,  $100-250 \mu m$  were determined. To determine mathematical functions describing (mass and particle number) changes in the fractional composition of each dust the results were statistically analyzed by applying the nonlinear estimation. Using of the polynomial regression provided a very good correlation between the sought functions and results of measurements. The results of measurements are presented in tables and charts. High degree of granulometric diversity of the investigated dust samples, depending on the dust formation mechanisms, i.e. on the technology used and composition of raw materials, was noted. High share of the fine particles in the dust may be a serious hazard to human health, especially in the case of breakdown in functioning of dust collectors. 24

## INTRODUCTION

Zinc and lead smelting creates potentially environmental hazards since large quantities of contaminants of high toxicity are produced in the technological processes. These contaminants include dust and heavy metals it contains, sulphur dioxide, hydrogen sulphide, carbon oxide, nitrogen oxide, sulphuric acid, aliphatic and aromatic hydrocarbons. Although the emission of the contaminants from technological processes of zinc and lead processing plants is not high in a global scale, it contributes to the local environment pollution significantly due to their toxicity. Zinc and lead metallurgy involves great variety of different technologies and raw materials. Thus, the physical-chemical properties of the contaminants produced are essentially different. Concentrations of these contaminants in smelter gases may also vary within very wide limits, even when the same technology is applied [8, 9].

In the past, the most important ecological problem related to zinc and lead smelting was  $SO_2$  emission to the atmosphere. At present, after having introduced efficient procedures of  $SO_2$  utilization by sulphuric acid production, the main problem is emission of heavy metals [1, 2, 4].

Dust released from technological sources and ventilation systems of industrial plants may become a serious risk for the atmosphere and other environmental components and in consequence for human health. The risk results from the fact that the dust particles are very fine and contain large quantities of very toxic elements (Pb, Cd, As, Tl) which may disturb the functioning of human organism. They may e.g. affect haematopoietic system or central nervous system or, in the case of cadmium and arsenic, cause cancer.

The extent of the harmful effect of dust on organisms depends on its fractional composition as well as on its chemical composition and mineralogical composition. The size of the particles determines the possibility of dust penetration to respiratory tract and release of the particles in different parts of the respiratory system. The most harmful are the grains whose diameters are smaller than 5  $\mu$ m since they penetrate pulmonary alveoli very easily. Moreover, specific surface of fine particles is usually large which favors their enrichment in many elements, including the toxic ones, in result of condensation.

Owing to high density (most often higher than 5 g/cm<sup>3</sup>) most of the dust emitted is deposited not far from the emission sources – at the distance of several or about a dozen kilometers. However, at favorable meteorological conditions, the finest dust fractions may be transported at long distances and, in this way, because undesirable results for long periods, far from the emission source.

Although the effect of the activities of heavy metal mining and processing plants on the natural environment has decreased in the last ten years [3, 7], dust emission from these sources is still a serious ecological problem.

#### MAIN SOURCES OF DUST EMISSION

Production of the zinc-works where the tests were carried out is based on the technology of simultaneous melt of zinc and lead in a blast furnace – Imperial Smelting Process (ISP). Metals obtained in this way are then refined or rectified. Lead refining plant is also equipped with a Short Rotary Furnace (SRF) in which secondary lead-bearing materials are processed.

Dust sources are found at every stage of the production process including transport, ore and charge preparation, heat and refining. The quantities and characteristics of dust

released from particular sources vary in a wide range, depending on the composition of raw materials processed, mechanism of the dust release, the process parameters and other less important factors. Dust release is the highest during Dwight-Lloyd process, in a blast furnace and when lead-bearing materials are melted in a SRF. Therefore, these sources have been equipped with bag filters of the efficiency exceeding 99.9%.

# CHARACTERISTIC OF THE DUST SAMPLES TESTED

The tests, measurements and analyses of the samples of dust were carried out in order to know fractional composition of technological and ventilation dust found in bag filters at different zinc-works plants [5]. The sampling points covered the sources characteristic of the technologies and essential when considering both the raw material quantity and dust quantity i.e. the sources which would affect the environment the most strongly if the dust collectors broke down. In order to get information on the fractional distribution of dust from other sources (e.g. blast furnace technological gases or dust sources where scrubbers were used) the samples from the sinter plant storage yard were analyzed. These samples were a mixture of all returnable materials directed to the sintering process.

Thus, the dust tested and analyzed was sampled at the following handling points of the technological process:

- Sinter plant crushing centre (12-chamber-filter servicing the sinter crushing and transfer); sample denoted P1.
- Sinter plant (12-chamber-filter operating at the area of the overflow of the collecting conveyor of the fit mix, mix discharge on to the sintering machine and the first-stage roll crusher); sample denoted **P2**.
- Blast furnace (14-chamber-filter covering the area of the slag and lead tapping, zinc vapor condenser and zinc refinement); sample denoted **P3**.
- Sinter plant (24-chamber-filter operating at the area of the hot sinter discharge from the sintering machine and the sinter pre-crushing); sample denoted **P4**.
- Lead refining plant (10-chamber-filter, servicing all the technological sources i.e. refining boilers, SRF, distillation furnace and muffle furnace); sample denoted **P5**.
- Sinter plant storage yard technological process products returned to the sintering process (dust, sludge, melting loss, slickers); sample denoted **P6**.

## **EXPERIMENTAL**

Seven series of the materials (each consisted of about 5 kg of dust, sampled every two or three months) were gathered for the analyses.

The dust samples were first dried at the temperature of  $105^{\circ}$ C and then screened through a sieve to remove larger grains (the mesh diameter was 250 µm). In most cases, the screenings were the sample part of minor importance except the dust samples P6, which contained a great number of particles of longer diameter, such as fast conglomerates, slag particles etc. However, since the range of such particles is not wide when they are introduced to the atmosphere, they were neglected in further analyses.

In the next stage, numerical fractional composition analysis of the samples was carried out with the use of a Laser Fractional Composition Analyzer Analysette 22 made by Fritsch and based on the phenomenon of Fraunhofer diffraction of He-Ne laser rays. A dust sample

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of 50–100 mg was placed in a beaker of 100 cm<sup>3</sup> and then alcoholized with about 50 cm<sup>3</sup> of isopropyl alcohol. Before the suspension was introduced to the apparatus, the samples were additionally subjected to ultrasonic disintegration to avoid measurement errors resulting from not complete dispersion of the agglomerates of dust particles. A doubled measurement range was used during the measurements, which made it possible to carry out the analyses in the range of dust particles size between 0.16 and 250  $\mu$ m.

In order to determine the mass fractional composition of the dust tested, sieve analysis (40–250  $\mu$ m) and sedimentation analysis (below 40  $\mu$ m) were applied.

During the sieve analysis, dry sample material of the mass of about 250–400 g was analyzed.

Before the sedimentation analyses of the dust samples, specific gravity of each sample was determined with the use of the air pycnometer for small samples. The sedimentation analyses were carried out by means of a standard Andreasen pipette. Isopropyl alcohol was used as the sedimentation liquid.

In result of the analyses, mass fractions of the following grain size intervals: 0-2.5; 2.5-5; 5-10; 10-20; 20-32; 32-40; 40-63; 63-100;  $100-250 \mu m$  were determined. In order to define a relation between the grain diameters and their fractional percentage, statistical analysis of the results of the fractional composition analysis was carried out. Different estimation methods were applied (linear, exponential and others). In result, the best fitting was acquired when describing the relation between the variables by a product of 5-degree polynomials. Then, the parameters of the assumed non-linear regression model were estimated in an iteration process with the use of quasi-Newton method. The model accuracy was evaluated by determining the value of the correlation coefficient R.

# The TEST RESULTS

The aim of the numerical fractional composition analysis was to get information about the actual refinement of the dust tested which was possible only by means of modern instrumental techniques. Table 1 shows the percentage of particles of different size in the samples tested.

The grain size interval, μm	Numerical percentage of the fraction, %							
	P 1	P 2	P3	P4	P5	P 6		
0-2.5	10.67	9.58	73.93	33.61	57.02	41.28		
2.5-5	6.71	7.32	15.69	12.06	19.50	13.13		
5-10	10.24	14.33	6.07	14.90	13.02	16.41		
10–20	20.77	27.50	3.32	21.14	7.76	18.10		
20-32	18.15	21.09	0.77	12.80	2.14	7.19		
32-40	8.89	7.73	0.16	3.28	0.35	1.85		
40-63	14.74	8.83	0.06	1.97	0.18	1.79		
63-100	6.80	3.13	0.01	0.23	0.02	0.23		
100-250	3.03	0.50	0.00	0.01	0.00	0.03		

Table 1. Numerical fractional composition of the dust samples tested (means values)

The tests have proved significant diversification of the numerical fractional composition of the samples tested. The percentage of fractions smaller than 10 µm is about 18–95%, whereas the percentage of particles larger than 100  $\mu$ m is between 0–3%. The differences in the fractional composition result mainly from the process of dust release and composition of the materials processed. Dust captured by the bag filters in the Sinter Plant (P1, P2 and P4) originates mainly from the process of the sinter mechanical crushing so it is dispersion dust. Dust from the sinter disintegrating plant and 12-chamber-filter of the sinter plant (P1 and P2) is released in result of crushing and dumping of the foundry mixture and sinter. Thus, the size of the dust particles depends, above all, on the brittleness and cleavage of the raw materials and the sinter, which are determined by the minerals they consist of. The composition of dust from the 24-chamber-filter (P4), in which the percentage of fine particles is high (about 60% of the particles smaller than 10  $\mu$ m), is affected mainly by the dust released at the discharge from the sintering machine. In the process the hot sinter cracks and volatile vapor of metals as well as very fine dust particles, found inside the sinter porous structure, are released. In the process of charge penetration in the blast furnace and in the process of lead refining, connected with evaporation and condensation, condensation dust is released (P3 and P5). The percentage of the particles smaller than 5  $\mu$ m is very high in this dust. Its larger particles are released in mechanical process occurring in the shaft of the IS furnace, in SRF and when these two furnaces are charged. P3 dust particles are the finest; the percentage of particles smaller than 1 µm is about 40% whereas the percentage of particles larger than 10 µm does not exceed 5%.

Fractional distribution of **P6** dust, being a mixture of waste products of all the technological processes (dust, sludge, melting loss, slickers) is a resultant of all the dust sources. More coarse fractions originate from the crushing and transport processes of the charge materials and products of particular technological handling points. They are also released from the processes in which very small particles are mainly released: zinc vapor condensation (melting loss from the pump well), slag and lead tapping or slag granulation.

Mass fractional compositions of the dust tested were also determined, which is the basis to define distributions of metals in dust [6]. Table 2 presents the mass percentage of the particles of different size in the dust samples tested.

The grain size interval, μm	Mass percentage of the fraction, %							
	P 1	P 2	P3	P 4	P 5	P 6		
0-2.5	4.99	5.88	40.61	20.38	33.31	32.03		
2.5-5	5.56	5.60	9.66	8.64	11.76	8.43		
5-10	10.86	9.83	11.27	10.37	12.33	9.88		
10-20	14.42	19.50	13.03	13.99	14.04	11.46		
20-32	10.35	14.87	6.38	10.00	7.72	6.06		
32-40	6.55	9.81	4.32	6.58	5.17	4.08		
40-63	23.84	28.93	10.59	18.13	12.33	10.50		
63-100	16.29	4.80	1.95	8.58	1.19	6.42		
100-250	7.14	0.79	2.19	3.32	2.15	11.14		

Table 2. Mass fractional composition of the dust samples tested (mean values)

The analysis shows that the dust tested agglomerated to different extents. The agglomeration was particularly apparent in the case of dust from the lead refining plant (P5). Its degree was smaller for the dust from the blast furnace, 24-chamber-filter of the sinter plant and the sinter plant storage yard (P3, P4 and P6). Dust captured by the 12-chamber-filter of the sinter plant (P2) formed very few fast agglomerates whereas no agglomerates were observed in the case of the dust from the sinter crushing plant (P1).

The fractional composition is strongly affected by the occurrence of fast agglomerates. Above all, the percentage of the particles larger than 100 µm is higher than it could be expected based on the numerical distribution of the fractional composition. It is a result of the phenomenon of occlusion (clinging, adhering) of the larger particles by the finer ones (in which finer particles cling to the larger ones). The highest percentage of the 40–63  $\mu$ m fractions (about 24–29%) was observed for the dust samples P1 and P2. In the other samples 0-2.5 µm fraction prevailed its percentage varied from about 20% for P2 to more than 40% for P3. These results show that when dust is released during high temperature processes (P3, P5, partly P4 and P6) the effect of evaporation and condensation processes dominates, whereas for the other dust samples the effect of dispersion is the most essential. Dust from the 24-chamber-filter (P4) is a mixture of condensation particles, released mainly when the hot sinter is discharged from the sintering machine, and dispersion particles from the processes occurring at the sinter flux transport and handling. Dust P6 is a mixture of particles from numerous sources, both from condensation and dispersion processes. The high percentage of 100–250 µm fraction may show that fast agglomerates are formed in result of changes caused by atmospheric factors at the storage yard.

Mean distributions of the numerical fractional composition and mass fractional composition of the dust tested are shown in Fig. 1 and Fig. 2.

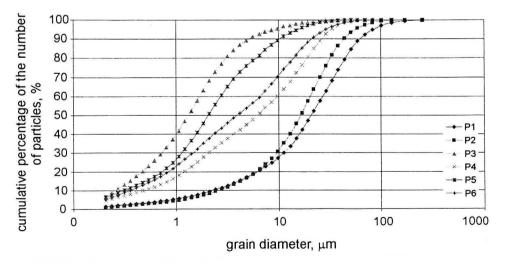


Fig. 1. Cumulative curve of numerical fractional composition of the dust tested - mean values

#### GRANULOMETRIC COMPOSITION OF DUST ...

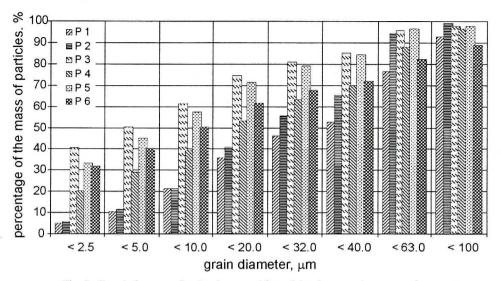
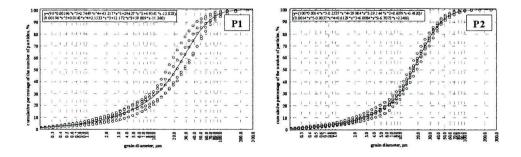
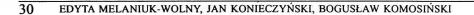


Fig. 2. Cumulative mass fractional composition of the dust tested - mean values

In order to find mathematical functions describing the changes in the fractional composition of the dust tested, statistical analysis of the results obtained was carried out by means of the non-linear estimation method. Application of polynomial regression model ensured good correlation between the functions and the measurement data. The correlation coefficient values varied between 0.985 and 0.998 for numerical fractional compositions and between 0.968 and 0.995 for mass fractional compositions while the extreme values were found for **P5** and **P2** dust samples. The statistical analysis made it possible to determine fractional composition for randomly selected grain size intervals within the range analyzed. Thus, the limitations of the sieve and sedimentation analysis were eliminated. It is of particular importance, when calculating the quantity of the mass flux of the pollutants released from the source or when calculating the emission of particular dust fractions if the interval efficiencies of the dust collectors used are known. The results acquired are illustrated in Fig. 3 and 4.



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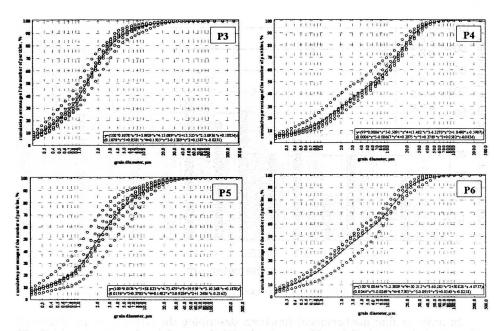
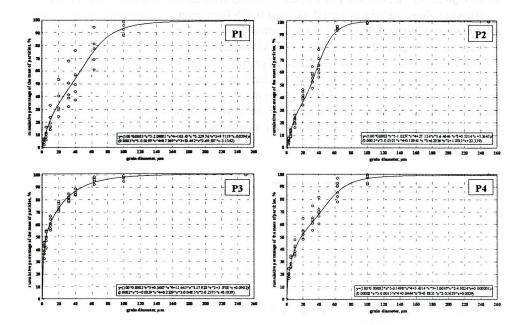


Fig. 3. Measurement results of the numerical fractional composition of the dust tested, the regression curves and the relevant regression functions



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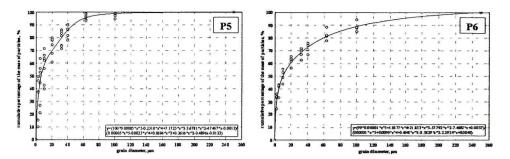


Fig. 4. Measurement results of the mass fractional composition of the dust tested, the regression curves and the relevant regression functions

#### CONCLUSIONS

- 1. The tests carried out have confirmed a significant differentiation of the fractional composition of dust released at particular handling points of zinc and lead smelting.
- The differences in the fractional composition of dust from particular source result, above all, from mechanisms of dust release depending on the conditions in which the technological process is carried out and composition of the raw materials processed.
- 3. Dust released from tested sources particularly from condensation processes since the high contents of respirable particles may be hazardous for the population when dust collectors break down.

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Received: July 5, 2005; accepted: January 25, 2006.

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