Size Distribution of Particulates Emitted from a Horizontal Spike Soderberg Aluminum Reduction Cell

Thomas R. Hanna and Michael J. Pilat
Water and Air Resources Division, Department of Civil Engineering, University of Washington

Aerosol size distributions were measured in the air exhausted from a horizontal spike Soderberg aluminum reduction cell at the Kaiser Aluminum and Chemical Corporation plant in Tacoma, Wash. The particle size distributions were measured with the University of Washington cascade impactor, developed specifically for source testing. The particle mass concentrations and size distributions were found to vary significantly with changes in the cell process operations. For a typical aerosol size distribution at the exit of the cell hood the mass mean particle diameter was 5.5 microns and the particle size standard geometric deviation was 25.

There has been very little detailed information reported regarding the nature of the particle emissions from aluminum smelters. Because this information should be valuable in designing the required control systems, the University of Washington initiated a research project at the Kaiser Aluminum and Chemical Corporation plant in Tacoma, Wash. The purpose of the project was to characterize the properties of the aerosols emitted from a horizontal spike Soderberg aluminum reduction cell as installed and operated at Kaiser's Tacoma plant. The aerosol characteristics measured included:

1. The aerosol size distribution as a function of the cell operating conditions, the cell air collection flow rate, and the distance down the air collection ducting from the cell.
2. The benzene soluble mass fraction of the particulates as a function of particle size.
3. The condensible portion (condensing between temperatures of 290 and 50°F) of the particulate mass concentration.
4. The fluoride percentage by mass of the particulates as a function of particle size.

Sampling Site and Conditions

Most of the aerosol samples were obtained from a single horizontal spike aluminum reduction cell located on potline 4 at the Kaiser Tacoma plant. A cross section of the cell is shown in Figure 1. Some features of this horizontal spike Soderberg aluminum reduction cell are:

1. The physical dimensions of the reduction cell are approximately 20.5 ft long by 13 ft wide by 8.5 ft high (outside dimensions).
2. The carbon anode paste (in a highly viscous form) is periodically fed into the anode casing (open at the top and bottom) where it is slowly baked and solidified as the anode is lowered within the cell and consumed in the reduction process.
3. The gases and particulates appear to be emitted mainly from the solidified crust area with two mechanisms of airborne particle formation present:
   a. Particles of the raw materials (mostly aluminum and cryolite) are entrained in the air when the solidified crust is worked (such as crust-breaking and aluminum siphoning operations) and during "flex-raising" (or when the flexible current carriers are manually raised to the next higher row of anode studs) operations.
   b. Volatilized materials from the molten electrolytic bath material escape when gas vents are present in the crust-forming particles in the colder air temperatures. These gas vents also entrain particles from the crust area. At least one such gas vent appeared to be present in the electrolytic crust nearly all the time although there were conditions when the complete crust was solidified.

   Some aerosols are also emitted from the sides of the anode because tars and hydrocarbons from the anode paste are forced from the anode during the baking process often catching on fire upon emerging from the anode. Carbonaceous particles are thereby formed during this combustion process.

Reprinted from APCA JOURNAL, *Vol. 22, No. 7, July, 1972*

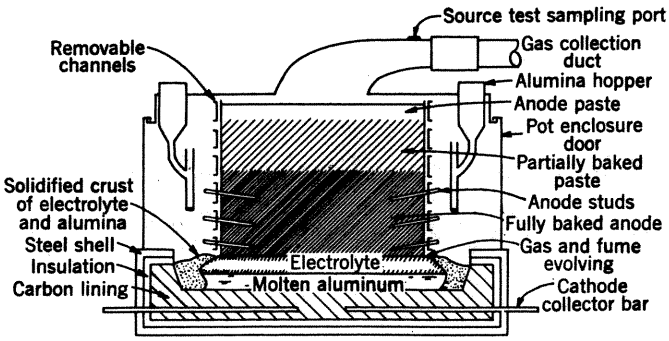

Figure 1. Cross-section of horizontal spike Soderberg aluminum reduction cell.

4. The entire pot is hooded and evacuated by two air collection ducts which transport the collected gases to a gas treatment process.
5. The cell maintenance doors are only open during periods of inspections, anode work, metal siphoning, and crust breaking. The doors remain closed during the rest of the time (which accounts for 95% of the total operating time of the cell and incluces the ore-feed operations which are automatically accomplished approximately every 30 min).

The approximate amounts of raw materials required for the production of one ton of aluminum (and which are added into the reduction cell) are shown in Table I.

Aerosol Particle Sampling and Analysis

The size distribution of the particulates emitted by the aluminum reduction cell was measured with a Mark II University of Washington source test cascade impactor. The U of W cascade impactor was developed in 1968 specifically for source testing. The theory, design, and performance of this device has been reported by Pilat, Ensor, and Bosch.[2] This cascade impactor is also being used in several other research projects concerning the emissions of particulate air pollutants from pulp mills, aluminum reduction plants, plywood veneer driers, fluidized bed sewage sludge incinerators, and hog fuel boilers. A Mark III model of the U of W impactor (patent pending) is commercially available under a licensing agreement with the University of Washington.[4]

The Mark II U of W cascade impactor consists of a series of 7 air jet stages. Directly below each jet stage is a particle collection plate on which the particle may impact and thus be collected. Those particles too small to impact upon the plates are collected on a filter located within the impactor downstream of the last collection plate.

Microscopic analysis (both optical and electron) of the particles collected on the plates was performed to verify the theoretical predictions of the University of Washington cascade impactor. The microscopic results conformed sufficiently with the calculated predictions to justify using the theoretical calibration equations.

The Mark II U of W source test cascade impactor is shown in Figure 2 together with the jet stages and the collection plates. A set of collection plates with a typical deposit of particles from an aluminum reduction cell is shown in Figure 3.

The procedure for using the source test cascade impactor for measuring the aluminum reduction cell effluent consists of three phases:

1. Pretest preparation, which includes cleaning the impactor, placing a thin layer of grease (Dow Corning high vacuum silicone grease) on the impactor plates (only necessary if solid particulates are to be sampled), and weighing the plates and filter (usually to the nearest 0.0001 gram).
2. Source testing, which involves determining the gas velocity profile in the duct, calculating the nozzle size for isokinetic sampling, and setting up the sampling train shown in Figure 4. Water condensation problems (which for the aluminum reduction cell emissions were minimal) were prevented by preheating the source test cascade impactor for approximately 2–5 min, prior to sampling. The particulate sample was then obtained, with typical test times of 4–10 min, and typical gas sampling rates of 0.6–1.4 scfm.
3. Analysis of source test results, which consists of disassembling the impactor and weighing the plates, computing the delta weight for each plate and the filter, and then inserting the results into a computer program to determine the size distribution for the source test.

The above source test procedure was used for determining the characteristics of the particles at duct conditions, and therefore no analysis was done on the contents of the Greenburg-Smith impingers. To determine the amount of hydrocarbons and condensibles present in the aluminum reduction cell emissions, four source tests were conducted with a sampling train including two Greenburg-Smith impingers filled with approximately 175 ml of carbon tetrachloride followed by a dry G-S impinger and two wash bottles containing ethyl alcohol (to collect the CCl_4 vapors). Additionally, the impactor plates were not coated with the silicone grease because the grease would interfere with later analysis. The particulates collected within the impactor were washed with benzene to determine the benzene-soluble portion of the particulates above 290°F, and the contents in the two impingers were analyzed by infrared spectroscopy for hydrocarbon content (as C–H) and for total condensibles (particles formed between 290° and 50°F) by evaporating the carbon tetrachloride and weighing the residual material (final drying took place at 400°F for approximately 5 min).

Table I. Raw materials for the production of one ton of aluminum in a Soderberg aluminum reduction cell.[1]

Material	Amount (tons)
Sulfur (impurity)	0.02
Alumina	1.9
Cryolite	0.04
Aluminum Fluoride	0.04
Fluorspar	0.003
Anode	
Petroleum Coke	0.47
Pitch Binder	0.14
Total: About 2.6 tons raw material per ton aluminum	

Results

A total of 140 particle size distribution tests were conducted. A composite size distribution of the particles emitted from the aluminum reduction cell (at temperatures above 290°F), presented in Figure 5 provides an indication of a typical size distribution (measured at the exit of the cell hood, the location shown in Figure 1). This size distribution is the result of combining all the size distribution data and taking into account the percentage of time each cell operation takes during a day. The significant features of this composite distribution (measured at a cell air collection flow rate of 1800 scfm) are:

1. The particle size distribution was quite polydisperse (the geometric standard deviation is 25);
2. The mass mean particle diameter was 5.5μ;
3. 30% by mass of the particles was less than 1μ in diameter;
4. 16% by mass of the particles was less than 0.2μ in diameter.

The particle mass concentration and size distribution were found to vary significantly with changes in the cell process operations. These variations are shown in Figure 6 for a typical process day (processes included are side crust breaking, end crust breaking, ore feeding, cell inspection, and aluminum siphoning). Some interesting features of these results (measured at an air collection flow rate from the cell of 1800 scfm) are:

1. The crust-breaking, aluminum siphoning, and ore-dump operations produce emissions of high particulate concentrations (average concentrations between 0.5 and 0.6 grains/scf), but the operations only last for short time periods of about 2-4 min. However, during these periods large amounts of visible airborne particulates appear to escape into the cell building (those aerosols escaping from the cell hood were not measured in this study).
2. Changes in the condition of the crust over the molten aluminum resulted in large variations in the particle emission characteristics. Between ore feeds and when the crust had no gas vents the particle mass concentration was low (about 0.05 gr/scf), the mass mean particle diameter was high (about 10μ), and the size distribution was very polydisperse (size geometric deviation greater than 1000). The samples with the large size geometric deviations typically had 50% of the sample mass on the first particle collection plate (particles of diameter greater than 12μ), about 30% on the filter (particles of diameter less than 0.2μ), and the remaining 20% on the other collection plates. When the crust was unstable and several gas vents were present the particle mass concentration was higher (about 0.12 gr/scf), the mass mean particle diameter was lower (in the 1-4μ range), and the particle size was less polydisperse (size geometric standard deviation of about 70).

When the air collection flow rate from the cell was increased from 1800 to 3900 scfm per cell, the average particle mass concentration remained about the same but the mass mean particle diameter increased from 5.5μ to approximately 12μ.

Source tests showed that the fraction of 0.5μ diameter particles 860 ft down the air collection duct from the reduction test cell was 50% less than at the cell exit. These data suggest that the small particles were coagulating in the duct. However, diffusion of the particles to the duct wall and settling of particles on the bottom of the duct may have been also occurring, thereby accounting for part of the reduction (qualitative measures of both these occurrences were made). It should be emphasized that the particulate size distribution 860 ft away from the test cell was made up of inputs from half of the reduction cells in potline 4. Therefore this size distribution is only indicative of what a particle size distribution 860 ft away from the test cell would look like (because it was made up of reduction cell inputs ranging from approximately 200 to 1500 ft away from the test point). Therefore no attempts were made to calculate particle coagulation coefficients.

Three sets of impactor collection plates with their deposited particles were analyzed for total fluoride content

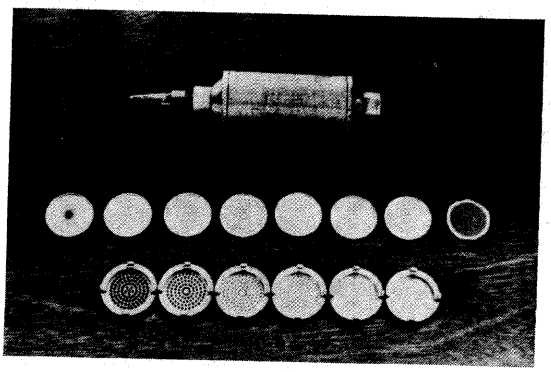

Figure 2. Mark II U of W source test impactor, showing (1) isokinetic nozzle, (2) cylindrical body, (3) filter holder and end of impactor, (4) collection plates, and (5) jet stages.

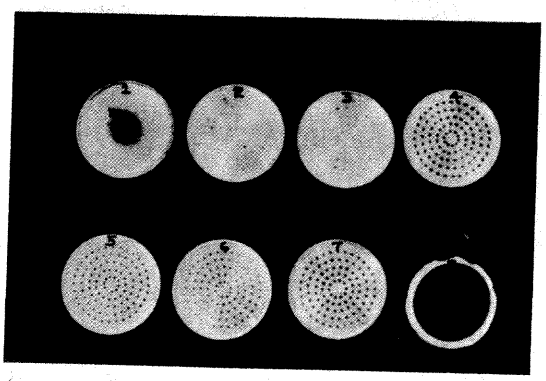

Figure 3. Set of impactor collection plates with a typical aluminum reduction cell particulate deposition (Number on plates indicate their order in the impactor).

by Food and Chemical Research Laboratories in Seattle, Wash., with the specific ion electrode method. Approximately 50% of the particulate fluorides by weight was associated with particles greater than 13μ in diameter (the particulate fluoride measured as F). Additionally, less than 10% of the total particulate sample by weight (and above 290°F) was fluoride (as F).

The results of the benzene-soluble, hydrocarbon, and condensible tests were:

1. The benzene-soluble portion found on the impactor plates for all four tests amounted to approximately 8–15% of the total weight of particulates on the impactor plates and filter. These particulates are probably made up mainly of hydrocarbons. The actual size distribution of these particulates was determined only approximately (because no silicone grease was applied to the impactor plates), with the results that the particles appeared to have a standard geometric deviation of 25 and a mass mean particle diameter of approximately 10μ.
2. The amount of condensible and/or particulates passed through the filter amounted to approximately 15% of the total particulat weight. When the CCl_4 had been evaporated off, the residual material was yellowish in color and was viscous in nature.
3. The amount of hydrocarbons (as C–H) detected in the CCl_4 of the impingers ranges between 3 and 6% of the weight collected in the impactor.

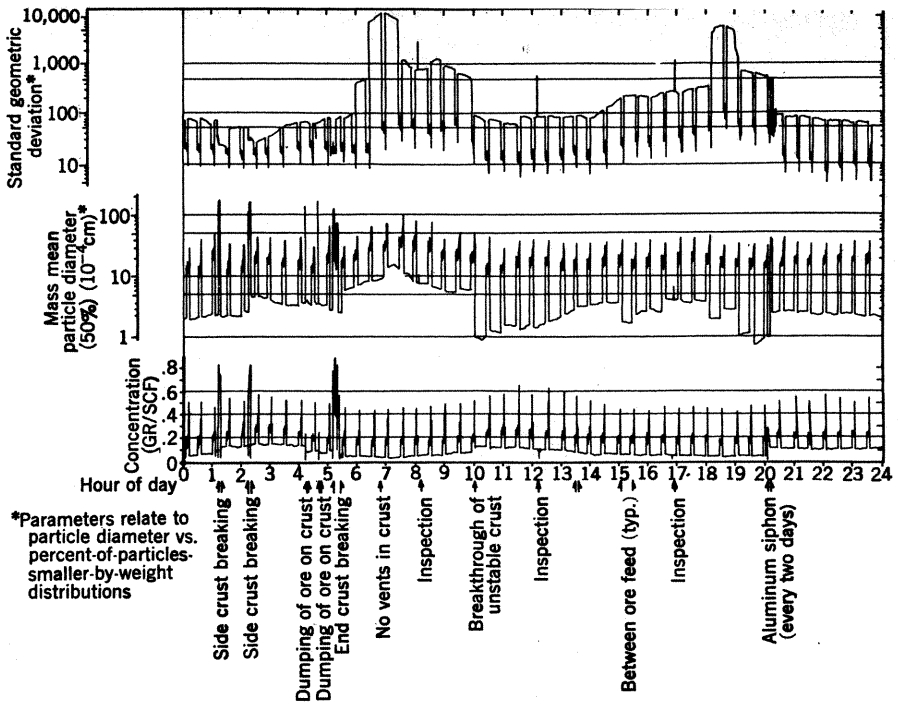

Figure 6. Particle airborne emissions as functions of daily process operations, at the exit of the aluminum reduction cell (air collection flow rate = 1800 scfm, air collection temperature = 290°F).

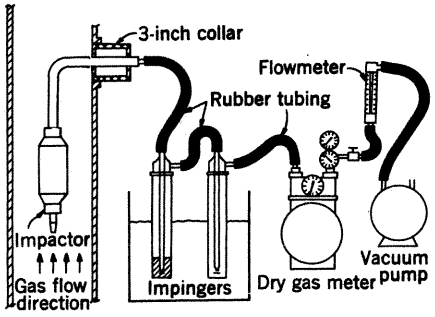

Figure 4. Schematic of sampling train used to test for particulates in the aluminum reduction cell effluent.

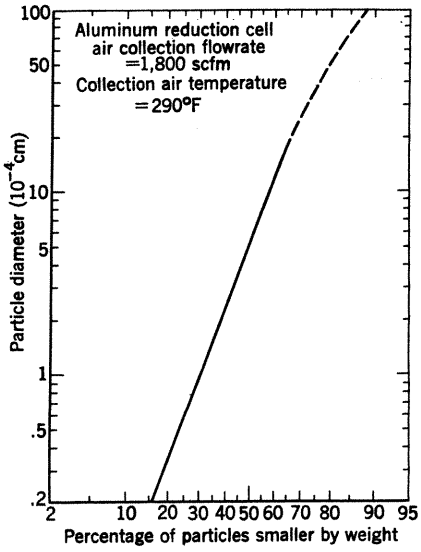

Figure 5. Composite particle size distribution by weight for aluminum reduction cell air emissions (Kaiser Aluminum & Chemical Corporation Plant at Tacoma, WA, potline 4), at the exit of the reduction cell.

Acknowledgments

This research was partially supported by the U. S. Public Health Service, National Air Pollution Control Administration Training Grant No. AP-29. The assistance of and discussions with David S. Ensor, Al Wendel, John Thielke, Robert J. Charlson, and August T. Rossano are gratefully acknowledged. The helpful assistance of Kaiser personnel including Dick Humphreys, Karl Klason, Hal Pearson, Gerry Brieger, and Chuck Schimmel and the cooperation of the Kaiser Aluminum and Chemical Corp. is most sincerely appreciated. The assistance of Bruce Smith of the Boeing Quality Control Laboratories with the analysis of the hydrocarbons in the CCl_4 is gratefully acknowledged.

References

1. J. Block and S. Moment. *Pacific Northwest Economic Base Study for Power Markets—Aluminum.* U. S. Dept. of Interior, Bonneville Power Administration, Vol. II, Part 7B (1967).
2. M. J. Pilat, D. S. Ensor, and J. C. Bosch. "Source test cascade impactor," *Atmos. Environ.*, **4**, 671 (1970).
3. T. R. Hanna, *Characteristics of the Airborne Aerosol Emissions from a Horizontal Spike Aluminum Reduction Cell.* Master of Engineering Thesis, University of Washington (1970).
4. University of Washington and Battelle Development Corporation, License for commercial sale of University of Washington Source Test Cascade Impactor to: Pollution Control Systems Corp., 321 Evergreen Bldg., Renton, Wash. 98055 (October 1970).

Mr. Hanna is Air Quality Control Program Supervisor, Dept. of Environmental Conservation, Juneau, Alaska, 99801 and Dr. Pilat is an Associate Professor of Air Resources Engineering, Water and Air Resources Engineering, Water and Air Resources Division, Department of Civil Engineering, University of Washington, Seattle, WN 98195. This paper was presented as Paper No. 70-AP-12 at the 1970 Annual Meeting of the PNWIS-APCA, November, 1970.