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Relationship of Plume Opacity to the Properties of Particulates Emitted From Kraft Recovery Furnaces

Abstract: The theoretical and measured relationships between the plume opacity (one minus the fraction of light transmitted through the plume) and the properties of the particulates emitted from a kraft recovery furnace are compared. The plume opacity (Ringelmann number) was measured with a Bailey smokemeter. The particle size distribution was analyzed with a cascade impactor and the particulate mass concentration was measured with an alundum thimble. A linear regression analysis of the measured data showed that over the Ringelmann number range of 0.85 to 3.0 there is a nearly linear relationship between the logarithm of the plume light transmittance and the aerosol mass concentration. The effects of the particle size distribution, particle density, and the stack gas temperature on the correlation between the plume opacity and the particulate mass concentration are discussed.

Keywords: Sulfate pulping · Recovery furnaces · Chimneys · Exhaust gases · Plumes · Opacity · Smokemeters* · Particles · Physical properties · Particle size distribution · Density measurement

THE research objective was to establish a relationship between the plume opacity (measured inside the stack) and the properties of the particles emitted from a kraft pulp mill recovery furnace. The use of a smokemeter to monitor the particulate mass concentration is a promising concept, but its application depends upon the development of a consistent relationship between light transmittance and particulate mass concentration. This paper presents part of a feasibility study of using stack smokemeters to monitor recovery furnace emissions and includes the results of simultaneous measurements of plume opacity, particulate mass concentration, and particle size distribution.

Cooper and Haskell (*1*) and Gansler (*2*) reported the use of Bailey bolometers to measure chemical ash losses from a kraft mill recovery furnace. Gansler presented the light transmittance and the particulate mass concentration of the aerosol emissions at various black liquor burning rates in a recovery furnace and various power inputs to a wet bottom, vertical flow electrostatic precipitator. No discussion was provided to indicate whether the data scatter was due to measurement errors or to changes in the particle properties caused by different furnace and electrostatic precipitator operating conditions. Changes in the particle size distribution were reported by Mitchell and Engdahl (*3*) as a possible problem in developing in-stack light transmittance to particle mass concentration relationships.

RESULTS

The results of 18 tests of simultaneous measurements of particulate mass concentration, particle size distribution, and plume light transmittance are presented in Figs. 1–4. The testing was done for 10 days during the period of June through August 1970. To provide a light transmittance range larger than that given by normal variations in furnace operation, the power to one or both of the two fields of electrostatic precipitator cell No. 3 was shut off during some of the tests. This provided a range in light transmittance measurements of from 83 to 41% transmittance.

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The measured light transmittance-particulate mass concentration relationship is presented in Fig. 1, which also shows the 90% confidence limits from a straight line regression analysis between the light transmittance and mass concentration measurements. In Fig. 2, the relationship is compared to the one reported by Gansler (*2*). To be able to directly compare the relationships, Gansler's data were normalized with the Beer-Lambert law to the 3-ft path length used at our sampling site since the light path length of the smokemeter used by Gansler was 13½ ft. The relationships are quite similar but it is apparent that results obtained at one specific recovery furnace are not generally valid for the emissions from other furnaces. Particle properties will usually vary between furnaces because of differences in furnace and gas cleaning equipment design.

The cumulative particle size distributions by mass measured with a cascade impactor are presented in Fig. 3, which includes three average particle mass distributions at full power to the electrostatic precipitator (12 tests) with one precipitator field shut off (4 tests) and with both precipitator fields off (2 tests). The seven data points of each distribution are the calculated d_{50} diameters and the cumulative fractional weights of each of the seven impactor stages. Table I shows the size distribution parameters of the three average distributions.

Bosch (*4*) measured the particle size distribution from the same recovery furnace with a cascade impactor. The sampling site was located downstream from the fan shown in Fig. 5. He reported an average particle size distribution for the combined emissions from all three precipitator cells of 1.5 μm geometric mass mean diameter and 5.5 geometric standard deviation.

A comparison between the measured and theoretically calculated relationships of the light transmittance to the particle mass concentration is presented in Fig. 4. The particle mass concentration was calculated (see Eq. 7) from the measured light transmittance, the magnitude of K obtained from Figure 6, and a log-normal distribution approximation of the measured particle size distribution.

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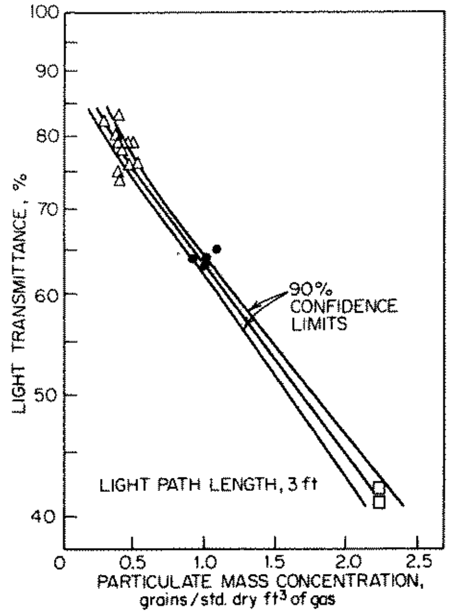

Fig. 1. Relationship of light transmittance to particulate mass concentration: (△) electrostatic precipitator on; (●) one electrostatic precipitator field off; (□) both electrostatic precipitator fields off.

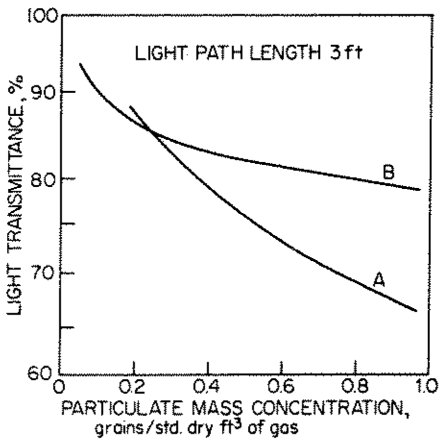

Fig. 2. Comparison of relationships of light transmittance and particulate mass concentration at two different kraft recovery furnaces: (A) this study; (B) from Ref. *2*. Illumination path length normalized to 3 ft.

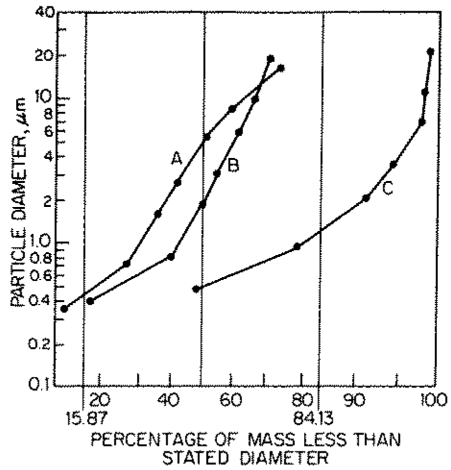

Fig. 3. Average particle size distributions of emissions: (A) electrostatic precipitator on; (B) one electrostatic precipitator field off; (C) both electrostatic precipitator fields off. Assumed particle density, 2.5 g/cm³.

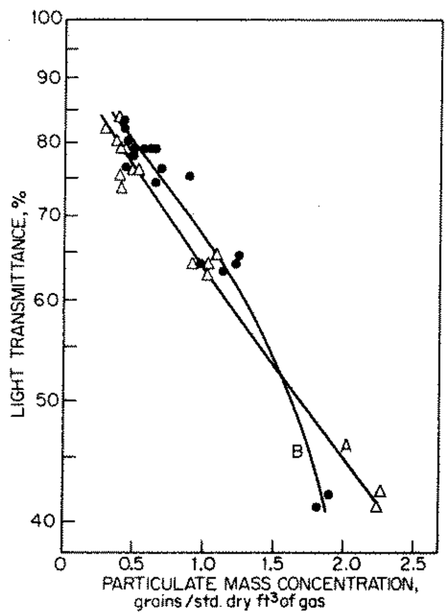

Fig. 4. Comparison of measured and calculated particulate mass concentration as a function of light transmittance: (A) measured relationship; (B) calculated relationship assuming log-normal size distributions, particle density of 2.5 g/cm³, refractive index of 1.5, and spherical particles.

DISCUSSION

Relationship of Particulate Mass Concentration to the Plume Light Transmittance

The Beer-Lambert law expressed in terms of K (see Eq. 7) indicates a linear relationship between the logarithm of the light transmittance and the particulate mass concentration (provided the average particle density and the value of K do not change as the particulate mass concentration changes). The 18 tests of this study represent a random sample of operating conditions of a typical kraft recovery furnace. The total variation of the calculated K values for the 18 tests is from 0.80 to 1.20. In Table II, this variation is compared to the corresponding variations of the size distribution parameters.

The averaging effect of the highly polydisperse size distribution is shown in Table II. All measured size distributions fall very close to the part of the K plot in Fig. 6 where the curves are almost flat. The nearly straight line of Fig. 1 is an experimental verification that K is almost constant and demonstrates that even large changes in particle size distribution parameters do not necessarily have a significant effect on the particle mass concentration-light transmittance relationship.

The comparison in Fig. 4 of the measured particulate mass concentration with that theoretically calculated using Mie light scattering theory and the measured particle size distribution shows good agreement. The theoretical calculation is based on the assumptions of log-normal particle size distributions, spherical particles, particle density of 2.5 g/cm³, a particle refractive index of 1.50, and a monochromatic smokemeter light source of wavelength 1.20 μm. In the light transmittance range from 63 to 83%, the theoretically calculated particulate mass concentration averages 28% higher than the measured value as given by the regression line. Below about 53% light transmittance, the calculated particle mass concentration is lower than the regression line of the measured values. The differences between the measured and theoretically calculated relationships may be caused by a number of factors. One possible cause is the inherent characteristics of smokemeters of optical design similar to the Bailey smokemeter to give a transmittance reading which differs from the theoretical value because the acceptance angle of the light detector is greater than zero degree. Ensor and Pilat (*5*) reported calculated factors for correcting the light transmittance measured at various acceptance angles. The correction in the light transmittance increases with decreasing magnitude of the light transmittance and thus may account for part of the discrepancy at the lower part of the curve in Fig. 4.

Statistical Analysis of the Data

The scatter of the data points around the regression line in Fig. 1 may be the result of both measurement errors and changes in the particle properties. A standard linear multiple regression analysis was performed on the measured data in order to determine the possible effects of the measured variables on the data scatter. The particulate mass concentration W was considered to be the dependent variable. The independent variables were: (1) logarithm of the light transmittance $\ln I/I_0$; (2) stack gas temperature, T_s (°F); (3) stack gas humidity, H_2O (% vapor by volume); (4) geometric mass mean diameter, d_{gw} (μm); and (5) deviation from isokinetic sampling, ΔR (%). The assumed mathematical relationship between the particulate mass concentration and the independent variables is

$$W = A \ln I/I_0 + BT_s + CH_2O + Dd_{gw} + E\Delta R + F + e$$

where

A–F = regression coefficients

e = the unexplained variation

Table I. Size Distribution Parameters

Electrostatic precipitator operation	*Geometric mass mean diameter, μm*	*Geometric standard deviation (approximate)*
Both fields on	5.0	10
One field off	1.85	13
Both fields off	0.5	5.5

Table II. Variation of K and the Size Distribution Parameters

Value	*Geometric mass mean diameter, μm*	*Geometric standard deviation*	*K, cm³/m²*
Maximum	11.0	17.1	1.2
Mean	3.6	10.3	1.0
Minimum	0.5	5.2	0.8
Maximum/minimum	22.0	3.3	1.5

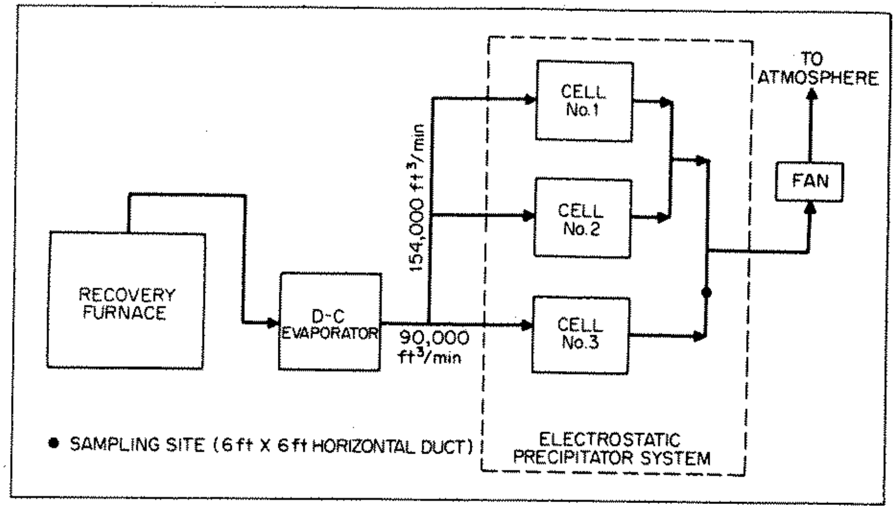

Fig. 5. Schematic of recovery system showing sampling site.

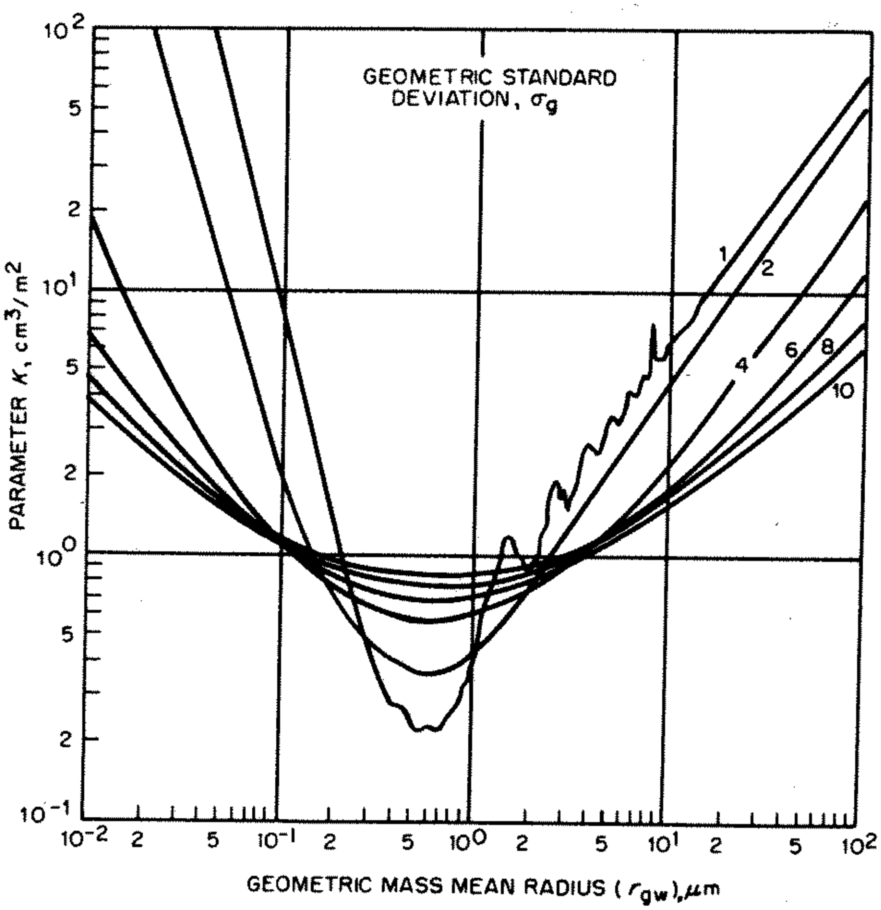

Fig. 6. Specific particulate volume/extinction coefficient ratio, K, as a function of log-normal size distribution parameters for an aerosol of spherical particles of refractive index 1.5 and at a radiation wavelength of 1.2 μm.

The results of the regression analysis are shown in Table III.

The changes in the light transmittance account for 98.2% of the total variation in the particle mass concentration measurements. The remaining 1.8% of the total variation represents the data scatter around the regression line of Fig. 1. Only a small amount of this 1.8% (20%) is explained by the variations in the size distribution represented by the particle geometric mass mean diameter. The variations in the stack gas temperature and humidity have an insignificant effect on the particulate mass concentration-light transmittance correlation. The remaining unexplained variation of 1.19% of the total variation in the dependent variable is probably due to errors in the measurement of the independent variables (light transmittance, particle size distribution, isokinetic flow rate, etc.). However, even with these errors, the 0.991 correlation coefficient between the light transmittance and the particle mass concentration is quite high.

THEORETICAL RELATIONSHIP OF PLUME LIGHT TRANSMITTANCE TO AEROSOL PROPERTIES

Opacity of Smoke Plumes

The light extinction property of an aerosol, as measured inside a duct or stack, can be expressed in terms of either % light transmittance T, % opacity O_p, or as a number on the Ringelmann smoke density scale. The relationship of the % light transmittance to the plume opacity is given by

$$T = 100 - O_p \quad (1)$$

The plume opacity measured inside the stack with a smokemeter is not necessarily the same as that outside the stack. The differences between the in-stack and out-of-stack plume opacities may be caused by differences in the plume dimensions (changes in the illumination path length through the plume) and by changes in the humidity of the stack gases resulting from cooling by the atmosphere. Increased relative humidity can cause condensation of water vapor and growth of the hygroscopic particles, thus increasing the plume opacity. The plume opacity as determined by a trained observer will depend on the ambient lighting conditions and the darkness of the background against which the white plume is observed. An extensive discussion of smoke plume optical effects and opacity observations was reported by Conner and Hodkinson (*6*).

Table III. Multiple Regression Analysis on Measured Data with Measured Particulate Mass Concentration as the Dependent Variable

Independent variables (in order of significance)	*Regression coefficient*	*Multiple correlation coefficient after variable was added*	*Variation caused by change in independent variable, %*	*Level of significance*
Light transmittance ($\ln I/I_0$)	2.73	0.9908	98.2	>0.99
Particle diameter (d_{gw})	−0.006	0.9926	0.36	>0.95
Deviation from isokinetic (ΔR)	−0.005	0.9936	0.19	>0.75
Gas temperature (T_s)	0.0016	0.9938	0.06	[a]
Water vapor (H_2O)	0.007	0.9940	0.03	[a]
Unexplained variation	...	...	1.19	
		Total variation =	100.00	

[a] Not significantly different from zero.

Light Extinction Theory

The transmittance of light through an aerosol can be described by the Beer-Lambert law

$$I/I_0 = \exp(-bL) \qquad (2)$$

where

I/I_0 = fraction of the incident light transmitted

b = light extinction coefficient

L = illumination path length

The light extinction coefficient b for an aerosol of polydisperse sizes ranging from radius r_1 to r_2 is given by

$$b = \int_{r_1}^{r_2} Q_{ext}(r,\lambda,m)a(r)n(r)dr \qquad (3)$$

where

Q_{ext} = light extinction efficiency factor of a particle of radius r and refractive index m at a light wavelength λ

$a(r)$ = projected area of the particle (πr^2 for a sphere)

$n(r)$ = particle number size frequency distribution function

The extinction efficiency factor can be calculated using equations theoretically derived by Mie (*7*) assuming spherical or other defined particle shapes.

Relationship of Light Transmittance to the Particulate Mass Concentration

The Beer-Lambert law can be written as a relationship between the light transmittance, I/I_0, and the particle mass concentration, W, by using the parameter K defined by Pilat and Ensor (*8*) as

$$K \equiv \frac{\text{total particle volume/volume of exhaust gas (cm}^3\text{/m}^3)}{\text{light extinction coefficient (m}^{-1})} \qquad (4)$$

$$K = W/\rho \qquad (\text{cm}^3/\text{m}^2) \qquad (5)$$

where

ρ = the average particle density, gm/cm³

W = the particle mass concentration, gm/m³

Substituting Eq. 5 for b into Eq. 2 gives

$$\frac{I}{I_0} = \exp\left(-\frac{LW}{K\rho}\right) \qquad (6)$$

Solving Eq. 6 for the particulate mass concentration W, we obtain

$$W = -\frac{K\rho}{L}\ln\left(\frac{I}{I_0}\right) \qquad (7)$$

The parameter K can be calculated using the Mie theory provided that the size distribution and shape of the particles are known.

Pilat and Ensor (*8*) reported calculations of K using number frequency size distributions (mathematical transformations of the log-normal distribution model from volume frequency size distributions to number frequency size distributions) and Mie theory.

Figure 6 presents the results of theoretical calculations of K as a function of the mass mean radius r_{gw} with the geometric standard deviation σ_g as a parameter for a particle refractive index of 1.5 and a wavelength of light of 1.2 μm. This refractive index was chosen to be representative of the kraft recovery furnace emission from handbook values of the pure compounds (*9*). The wavelength of light is an approximate average for the Bailey smokemeter light source. The parameter K may vary considerably with changing size distribution parameters. For polydisperse aerosols ($\sigma_g > 6$), however, the variation of K between r_{gw} of 0.1 and 5 μm is remarkably small. The wide range of particle sizes found in typical polydisperse particulate emissions tends to average out the optical scattering effects. This suggests that the average K for polydisperse sources in this size range (such as kraft recovery furnace emissions) will be within defined limits.

MEASUREMENTS OF LIGHT TRANSMITTANCE, PARTICULATE MASS CONCENTRATION, AND SIZE DISTRIBUTION

Sampling Site

Measurements of the light transmittance, the particulate mass concentration, and the particle size distribution were performed at the St. Regis Co. kraft pulp mill in Tacoma, Wash. The exhaust gases from a Combustion Engineering furnace with a rated capacity of 1.4×10^6 lb of dry solids per day pass through a direct contact evaporator and three parallel Research Cottrell electrostatic precipitator units before being discharged into the atmosphere. The sampling site was located downstream from the number 3 kraft recovery furnace as shown in Fig. 5. The sampling ports and a smokemeter were located about 50 ft downstream from the electrostatic precipitator unit number 3 and about 25 ft downstream from the nearest flow disturbance (a 90° bend). The gas velocity in the 6-ft duct ranged from 60 to 70 ft/sec and was relatively constant. The gas temperature ranged from 250 to 320°F and the water vapor concentration was about 25% by volume. The sampling port and the smokemeter are located close together so that these simultaneous measurements were representative of the same aerosol.

Smokemeter Measurements

The light transmittance was measured with a Bailey smokemeter. The light source was a narrow beam spot lamp (GE PAR 46/3NSP) and the light detector a sealed beam bolometer (GE 65HPAR/1). The average wavelength of light for this device was calculated to be about 1.2 μm. The light transmittance readings were recorded in Ringelmann numbers on a Bailey recorder. The illumination path length across a slotted tube between the light source and the bolometer was 36 in.

Particulate Mass Concentration Measurements

The particulate mass concentration was measured using a Western Precipitation alundum thimble holder with an RA-98 (coarse porosity) alundum thimble lined with glass wool. A 2-in.-diam. glass fiber filter was used to check the particle collection efficiency of the glass wool lined alundum thimble.

Analysis of Particle Size Distribution

The size distribution of the particles was measured with a University of Washington source test cascade impactor similar to that described by Pilat, Ensor, and Bosch (*10*). The cascade impactor includes seven multijet impaction stages followed by a 2-in.-diam. filter holder (for glass fiber filters or alundum filter disks) and was designed specifically for source testing. The impactor separates the particles into seven size classes ranging from about 0.5 to 20 μm, depending on the gas flow rate through the impactor and the mass density of the particles. The sampling train shown in Fig. 7 was used to obtain isokinetic mass concentration and size distribution samples.

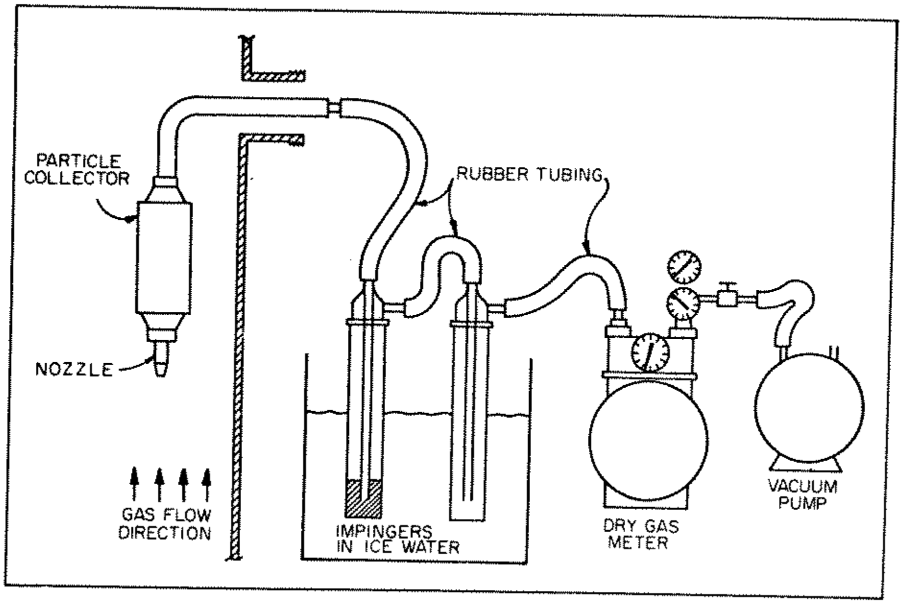

Fig. 7. Sampling train for isokinetic sampling.

Test Procedures

The following variables were measured in each test to characterize the stack gas properties: stack gas temperature, pressure, and velocity; gas meter pressure, temperature and gas volume metered; volume of water collected in the Greenburg-Smith impingers; and test duration. The following test procedures were used to minimize possible errors in the measurements:

(a) The smokemeter was calibrated with standard density grids several times during the study. The spot lamp and bolometer cover glasses were cleaned prior to each test.
(b) The thimble holder and impactor were inserted into the stack during the tests as shown in Fig. 7. The use of a straight inlet nozzle reduces particle losses in the nozzle. The probes were preheated for 10 min with the nozzles facing downstream in the stack prior to each test to avoid condensation of water vapor in the particle collectors.
(c) The sampling rate was carefully regulated to maintain nozzle inlet velocities as near to isokinetic conditions as possible. The stack gas velocity was measured before each test using an S-type pitot tube connected to a differential pressure gage. The sampling trains were checked for air leakage (which was found to be negligible at a vacuum of 15 in. of Hg).
(d) The procedure for preparation and weighing of the alundum thimbles, impactor plates, and glass fiber filters was consistent for each test. Before each test they were: (1) washed and cleaned; (2) dried (thimbles at 1100°F, plates and filters at 220°F); (3) cooled in a desiccator at least 12 hr; (4) weighed on a Mettler balance to 4 decimal points (to 0.0001 g); and (5) stored in clean petri dishes. After each test they were: (1) stored in laboratory environment overnight and (2) weighed on the same Mettler balance.

Calculation of Results

Standard equations reported by Haaland (*11*) were used to compute the stack gas velocity and the particulate mass concentration. The cascade impactor data were reduced with a computer program which calculated the particle diameter of a 50% collection efficiency, d_{50}, and the mass fraction of particles collected at each impactor plate. This actual measured distribution was fitted to a log-normal distribution using a least squares fit method with the data plotted as on logarithmic probability graph paper. The particle diameter at the 50% point is then the geometric mass mean diameter, and the geometric standard deviation σ_g is given by

$$\sigma_g = \frac{84.13\%\ \text{size}}{50\%\ \text{size}} = \frac{50\%\ \text{size}}{15.87\%\ \text{size}}$$

The geometric mass mean radius and the geometric standard deviation of this fitted straight line was used to obtain theoretical *K* values from Fig. 6.

CONCLUSIONS

A relationship has been developed between the plume opacity and the particle mass concentration measured downstream from an electrostatic precipitator collecting the particulates exhausted from a kraft recovery furnace. This relationship enables the determination of the particle mass concentration from the measured light transmittance within (90% confidence) a concentration of approximately ±0.05 grains/std. dry ft³ of gas at 0.3 grains/std. dry ft³ of gas and ±0.1 grains/std. dry ft³ of gas at 2.0 grains/std. dry ft³ of gas. The measured correlation of light transmittance to the particle mass concentration is in good agreement with a theoretical relationship which is based on Mie light scattering theory and particle size distribution measurements. A linear multiple regression analysis of our measured data has shown that variations in the particle size distribution (from 0.17 to 4.75 μm mass mean radius) have little effect (less than 0.36% of the total variation in *W*) on the relationship of the light transmittance to the particle mass concentration. This observed small effect of changes in the particle size distribution is in agreement with predictions based on theoretical calculations of a parameter *K* using Mie theory and the measured particle size distributions.

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