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ABSTRACT

For measurements of ore dust in uranium concentrating mills, a Sierra Model 235 slotted cascade impactor was calibrated at a flow rate of .21 m^3/min, using solid monodisperse particles and an impaction surface of Whatman #41 filter paper soaked in mineral oil. The reduction from the impactor's design flow rate of 1.13 m^3/min (40 cfm) to 0.21 m^3/min (7.5 cfm) increased the stage cut-off diameters by an average factor of 2.3, a necessary adjustment because of the anticipated large particle sizes of ore dust. The underestimation of mass median diameters, often caused by the rebound and reentrainment of solid particles from dry impaction surfaces, was virtually eliminated by using the oiled Whatman #41 impaction surface. Observations of satisfactory performance in the laboratory were verified by tests of the impactor in ore mills.

INTRODUCTION

In recent years, the validity of the special air concentration limit for uranium ore dust, established by the AEC in 1960 and later adopted by the NRC, has been questioned.[1,2] Animal inhalation experiments at Battelle Northwest Laboratories have indicated that the biological half-life of thorium-230 in ore dust is longer than the half-life of its parent, uranium.[3,4] This is contrary to the original assumption in deriving the concentration limit that the thorium-230 in uranium ore dust remains with uranium in the body, exhibiting the same 120-day biological half-life as uranium. Also, informal reports by various groups have shown occasional disequilibrium of uranium and its decay products in air samples of ore dust, whereas equilibrium was assumed in the derivation. Because of these uncertainties, the NRC is reevaluating the maximum permissible concentration (mpc) for uranium ore dust. The present investigation was undertaken to provide information about the physical characteristics of ore dust for use in the reevaluation. Cascade impactor samples are being collected in the ore processing areas of mills to determine particle sizes and radionuclide ratios.

Initial field tests were performed in several uranium mills with a 1 cfm Andersen cascade impactor to obtain approximate measurements of particle size and concentration. Many of the samples contained quantities of radionuclides that were too low for reliable chemical analysis. Also, activity median aerodynamic diameters (AMAD's) tended to exceed the $(D_p)0.50$* value of the first impactor stage, 11.0 μm. For more accurate measurements, a cascade impactor with larger cut-off diameters and capable of higher flow rates than the 1 cfm Andersen was needed. Sample volumes had to be on the order of 25 m^3, suggesting the use of a high-volume cascade impactor.

Among the available high volume cascade impactors, the Sierra Model 235 (Sierra Instruments, Inc., Carmel Valley, CA 93924) slotted five stage impactor was selected because of several favorable features and because a close familiarity with its characteristics had been acquired in a previous study.[5] Some deficiencies in its performance had been noted but they were thought to be correctable. For example, solid particles tended to rebound from the recommended glass fiber filter impaction surfaces, resulting in a downward shift of the apparent mass median diameter (MMD) of the sampled aerosol as larger particles were displaced to lower stages and to the final filter. In the course of the ensuing laboratory tests, substitution of Whatman #41 filter paper soaked in mineral oil eliminated rebound completely. The Whatman paper had the additional (and necessary) advantage of being free of radioactive contamination.

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*The diameter of a particle which will be collected with an efficiency of 50% by that stage.

Stage cut-off diameters were increased by a required factor of 2.3 by reducing the air flow rate to .21 m^3/min. Although much less than the design sampling rate of 1.13 m^3/min, .21 m^3/min yielded the required sample volumes in about two hours of operation, a reasonable collection period for the planned field study.

The impactor characteristics are given in Table 1.

THEORY

The Sierra slotted cascade impactor is composed of a series of five impaction stages followed by a filter. Each stage consists of parallel jets and a collection surface as shown in Figure 1. As aerosol-laden air passes through the jets, individual particles that have gained enough momentum will leave the airstream lines and impact on the collection surface, while those with insufficient momentum will follow the airstream to the next jet (Figure 2a). Since the jet velocities increase with each succeeding stage, particles of progressively smaller aerodynamic size are sequentially impacted and collected. Particles which have inertial forces too small to be impacted on the final jet stage are collected on a final filter.

The impaction efficiency, E, is defined as the fraction of the number of particles of a known size which are removed from the carrier airstream after striking the impaction surface. In an ideal impactor all particles which strike the impaction surface should be collected. However, this is not always the case and many types of adhesive coatings and filter surfaces have been used by various investigators to increase the stage collection efficiency.[6]

The impaction efficiency for individual stages is usually designated in terms of an effective cut-off at a point where E is 50%. This cut-off is often expressed as a function of the Stokes number $(Stk\ 0.50)^{\frac{1}{2}}$ or of particle size $(D_p)0.50$.

The Stokes number, as defined by Fuchs,[7] is the ratio of the particle stopping distance, x_s, to the half-width of the impactor throat, W/2,

$$Stk = \frac{x_s}{W/2}$$

the x_s term is defined as

$$x_s = \frac{\rho_p C V_o D_p^2}{18\ \mu}$$

so that

$$Stk = \frac{\rho_p C V_o D_p^2}{9\ \mu W}$$

where

ρ_p = density of particle, g/cm^3

C = Cunningham slip correction

V_o = average jet velocity, cm/s

D_p = particle diameter, cm

μ = fluid viscosity, poise

w = jet width, cm

and Stk is used as a measure of the dimensionless particle size.

Detailed flow fields in both rectangular and round impactors have been determined by Marple through numerical solution of the Navier-Stokes equation.[6,8,9] Using these flow fields and numerical integration, he determined particle trajectories and theoretical impaction efficiency curves for various values of jet-to-plate distance, jet Reynolds number and jet throat length.

Even though the theoretical performance of an impactor can be predicted accurately by numerical calculations, experimental results are found to deviate from predicted values if the experimental design does not correspond exactly to the numerical model. Experimental parameters such as inlet losses, interstage losses, particle rebound and reentrainment from impaction surfaces cannot be quantified on theoretical grounds and result in an S-shaped efficiency curve rather than the ideal curve as shown in Figure 2b. The position of the S-shaped curve relative to the ideal curve can be displaced either to the right or left depending on such factors as jet-to-impaction surface distance, Reynolds number, jet throat length, jet inlet design, and efficiency of particle collection.

Willeke, who calibrated the Sierra slotted impactor at its design flow rate of 40 cfm, also described the change in individual stage collection efficiency curves, when stage parameters and collection surfaces are varied.[10] His experimental data show that second stage collection efficiency curves are close to theoretical predictions when stage parameters approach ideal design conditions. However, significant shifts in collection efficiencies occur when the second stage is preceded by the first stage, which changes the ideal inlet flow conditions, or when parameters such as impaction surface and slot shape are varied.

Because of the foregoing reasons and since the proposed operating conditions differed significantly from those considered in any previous calibration of the Sierra impactor, an experimental calibration and determination of individual stage $(D_p)0.50$ values was deemed necessary.

EXPERIMENTAL APPARATUS AND PROCEDURES

Figure 3 is a schematic diagram of the test arrangement used for this calibration. The sampling chamber was maintained at a slightly positive pressure by controlling the amount of filtered dilution air which was added to the primary aerosol stream. This slight overpressure precluded ambient air particles from contaminating the test aerosol. The actual sampling time of each test run varied inversely with particle size being used, smaller particles requiring a longer sampling time in order to obtain an adequate amount of aerosol on the impactor stages for accurate analysis.

The principal aerosol used for this calibration consisted of solid, dry, monodisperse spheres of methylene blue in the aerodynamic size range of 1 to 20 micrometers. The particles were generated from water-alcohol solutions of methylene blue by means of a one-inch diameter variable speed, stainless steel spinning disc. Desired particle diameters were obtained by varying the disc speed and solution concentration. The solution feed rate was regulated by a Sage Model 351 syringe pump.

Particles of methylene blue smaller than 1 micrometer and greater than 20 micrometers were not adequate for calibration purposes. The smaller particles required extremely long sampling times in order to obtain quantities suitable for the spectrophotometric analysis method used for this calibration. Particles greater than 20 micrometers were usually non-spherical and had geometric standard deviations greater than 1.1. An attempt was made to extend the particle size range using uranine for smaller particles and iron oxide particles for larger particles. These tests were limited because of analytical problems, which were attributed to the incomplete removal of mineral oil from the leach solutions.

In preparation for use, the Whatman #41 filter paper impaction substrate was soaked in mineral oil until saturated, removed from the mineral oil, allowed to drip until all excess oil was removed, and then placed carefully on the impactor plate, preventing any of the mineral oil from entering the slots of the orifice plate.

Three impactors were run during each test, two impactors with oil-soaked Whatman #41 impaction surfaces and one with dry Whatman #41 impaction surfaces. The flow rate of each impactor was continually monitored by an inline orifice meter during test operations and confirmed by means of a positive displacement meter. The impactors were placed randomly in the three test positions within the sampling chamber during the test series.

ANALYSIS

At the completion of a test run, the slotted impaction papers were placed in separate, shallow 21 X 21 cm dishes and leached with ethyl alcohol. The final filter was leached in a similar manner.

Mineral oil was separated from the methylene blue-alcohol leach solution by centrifuging 35 ml of the solution in a conical tube for 15 minutes at 20,000 rpm. The supernatant liquid containing the methylene blue in alcohol was carefully decanted from above the precipitate which contained the mineral oil and any fibrous material recovered during the leaching process.

The methylene blue-alcohol solutions were read on a Turner Model 350 spectrophotometer at a wavelength of 525 nm. Solution concentration were determined from a calibration curve developed from known concentrations of methylene blue in ethyl alcohol. The total mass of each sample was the product of the concentration and volume of the leach solution.

Aerosol deposits on impactor surfaces, removed by thoroughly rinsing each stage after removal of the impaction filter, are presented as stage losses (e.g., stage loss consists of all particles collected on any surface of that impactor stage.) The areas of greatest loss, which became evident during rinsing, were in the orifice slots and the area between slots on the downstream face of the orifice plate. Some losses were also found on the upstream edges of the slots where the filter impaction paper occasionally failed to cover the plate completely up to the edge of the slot.

In the tests using particles other than methylene blue, uranine concentrations were determined by standard fluorometric methods and iron concentrations were determined by spectrophotometric measurement of the ferrous, 1-10 phenanthroline complex at 507 nm.

PARTICLE SIZING METHODS

A sample of input aerosol to the chamber was collected on a Millipore filter during each test. The collected particles were photographed with a light microscope and sized using a Zeiss TGZ-3 particle size analyzer.

In calibrating instruments, the usual method of obtaining the aerodynamic equivalent size of a solid spherical test aerosol is to determine the geometric mean size by microscopic methods and to correct for particle density, if it differs from unity. However, reported experimental determinations of the density of particles formed by drying droplets of ethanol-water solutions of methylene blue, uranine, or a combination of the two are not consistent.(11,12) In air sampler calibrations by other investigators the particle density was assumed to be the same as the density of the bulk material from which the particle was generated.(13,14)

The methylene blue particles used for this calibration appeared to be solid spheres when examined in the light microscope. However, scanning electron micrographs revealed that in some tests the smaller particles were similar to sponges (Figure 4) and larger particles had hollow cores (Figure 5). To eliminate uncertainties about particle density in this calibration, the settling velocity of the aerosol was determined during each test by a horizontal tube method, and then converted to an aerodynamic equivalent diameter.(15)

CALIBRATION RESULTS

A composite of the stage collection curves developed from this calibration is shown in Figure 6. These curves were used to develop the efficiency curves for individual stages, shown in Figure 7, which define the stage $(D_p)0.50$ values. A comparison of the theoretical and experimental stage $(D_p)0.50$ values is given in Table 2.

The results indicate that the required characteristics for measuring ore dust were attained at the selected operating conditions. At the flow rate of 0.21 m^3/min, the stage $(D_p)0.50$ values were greater by an average factor of 2.3 than the values determined experimentally by Willeke at the design flow rate of 1.13 m^3/min. The use of Whatman #41 paper soaked in mineral oil, as impaction surfaces, essentially eliminated particle rebound. Also it was found that the total interstage loss was less than 10% for all particle sizes that were covered.

Tests of the impactor using dry Whatman #41 paper showed greater interstage loss and evidence of particle rebound, although at 0.21 m^3/min rebound was not as severe as in previous tests at 1.13 m^3/min using dry glass fiber filter impaction surfaces.[5] An example of rebound is shown in Figure 8. The three cascade impactors were run simultaneously. Very close agreement is seen between the two impactors having oil-soaked Whatman #41 impaction surfaces but significantly different results were obtained for the third impactor having dry Whatman #41 impaction surfaces. In the latter case, the 16.2 μm test aerosol penetrated to lower stages and deposited on the walls to a greater degree.

During calibration tests, greater aerosol deposition was noted beneath the ends of the slots than along the line of impaction in between. The uneven deposition is attributed to the ends of the slot acting as round orifices and thus having greater collection efficiency than the intermediate slot. This effect, along with the lack of true monodispersity of the test aerosols, accounts for the non-ideal S shape of the efficiency curves shown in Figure 7.

RESULTS OF FIELD MEASUREMENTS

A number of samples were collected in an ore mill with Sierra cascade impactors operated at the modified conditions in order to verify laboratory observations. Figures 9 and 10 are log probability plots of representative impactor samples taken in the ore crushing area. The activity median aerodynamic diameter (AMAD) is calculated from radiochemical analyses of the uranium-238 nuclide collected on each impaction surface and on the final filter. The AMAD's and geometric standard deviations are computed from lines of best fit obtained from a least squares analysis. In Figure 9, two sets of duplicate test results are shown for impactors having oiled Whatman #41 impaction surfaces. In general, agreement of AMAD values for duplicate tests was within 10%; somewhat larger deviations were associated with AMAD values greater than 10 μm.

Duplicate tests comparing oiled and dry Whatman #41 impaction surfaces are shown in Figure 10. For the dry impaction surface, the AMAD of 6.7 μm is significantly lower than the 10.7 μm value obtained from the oiled surface. This difference is attributed to particle rebound from the dry impaction surfaces and displacement to lower impaction stages.

CONCLUSIONS

The basic requirements for this investigation of occupational exposure to ore dust are sufficient sample volume to meet analytical requirements, low radioactive background in the sample substrate, impactor stage cut-off diameters appropriate for aerodynamic median diameters in excess of 10 μm, and minimal particle rebound and inter-stage loss.

When operated in accordance with modifications adopted for measurements of uranium ore dust, the Sierra Model 235 slotted cascade impactor performs satisfactorily. Laboratory tests showing satisfactory performance were reenforced by preliminary measurement in an ore mill.

At a flow rate of 0.21 m^3/min, cut-off diameters were increased by a factor of 2.3, in approximate agreement with theoretical calculations, although differences were sufficient to justify the experimental calibration. The cut-off diameters are about optimal for the expected range of particle sizes. At 0.21 m^3/min, the sample volume required for accurate radiochemical analysis can be obtained in about 2 hours. Use of Whatman #41 filter paper soaked in mineral oil eliminated the radioactive background associated with glass fiber filter impaction substrates and eliminated particle rebound as well. Furthermore, wall loss was reduced to less than 10% of the collected sample.

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TABLE 1

CHARACTERISTICS OF THE SIERRA MODEL 235 HIGH-VOLUME SLOTTED CASCADE IMPACTOR*

Stage No.	W (cm)	T (cm)	S (cm)	T/W	S/W	L (cm)	No. of Slots	Slot End	V (cm/sec)	Re
1	0.397	0.635	0.317	1.6	0.8	12.4	9	round	80	420
2	0.163	0.114	0.186	0.7	1.1	12.4	10	rect.	175	380
3	0.091	0.114	0.186	1.2	2.0	12.4	10	rect.	312	380
4	0.046	0.114	0.186	2.5	4.0	12.4	10	rect.	618	380
5	0.025	0.114	0.186	4.6	7.4	12.4	10	rect.	1137	380
6	0.163	0.318	-	1.9		12.4	10	rect.	175	380

*Operated at 0.21 m^3/min

W = Slot Width
T = Throat Length
S = Jet to Plate Distance
L = Slot Length
V = Jet Velocity

$$Re = \rho \frac{V2W}{\mu}$$

$\rho = 1.2X10^{-3}$ g/cm^3
$\mu = 1.81X10^{-4}$ poise

TABLE 2

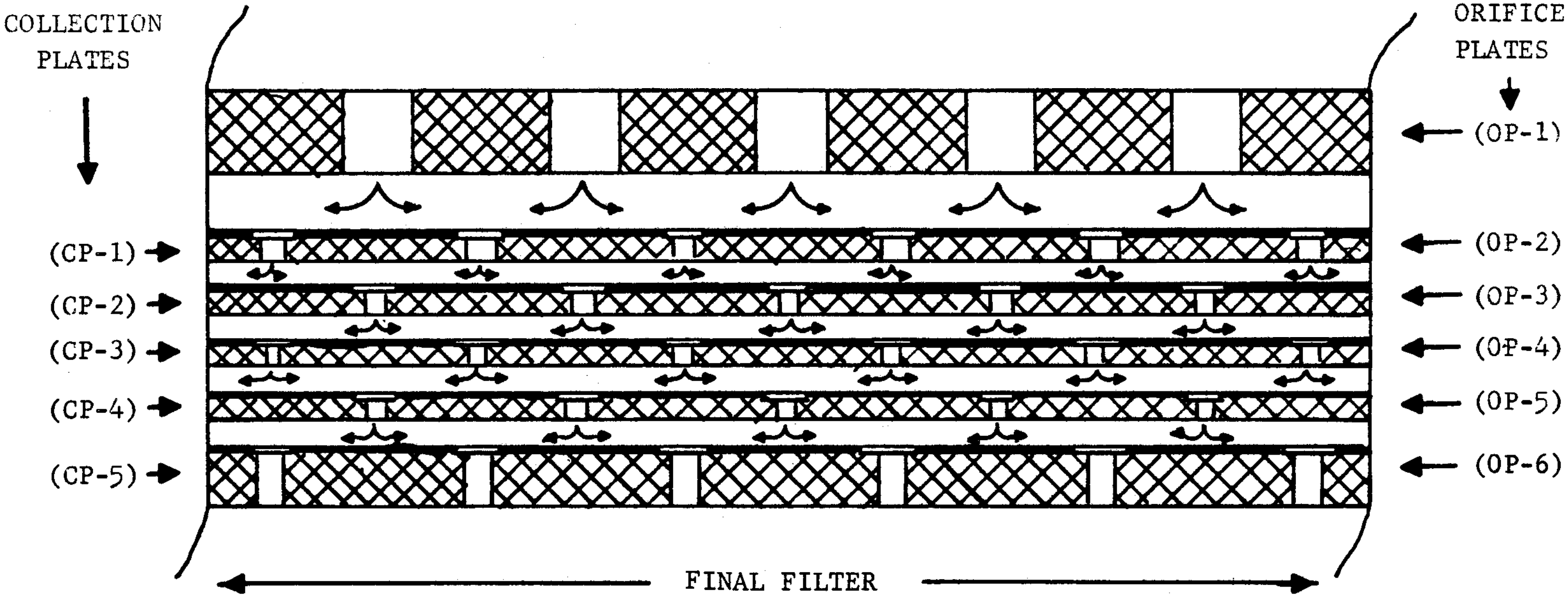
IMPACTION CHARACTERISTICS OF THE SIERRA MODEL 235 SLOTTED CASCADE IMPACTOR

Stage	$(D_p)0.50$ at 0.21 m^3/min (μm) Theoretical[1]	Experimental[2]	$(D_p)0.50$ at 1.13 m^3/min (μm) Theoretical[3]	Experimental[4]	Ratio: Exp. 0.21 m^3/min / Exp. 1.13 m^3/min
1	20.02	15.55	8.40	7.22	2.15
2	8.63	6.65	3.60	3.00	2.21
3	4.79	3.95	2.00	1.50	2.63
4	2.38	2.05	.97	.93	2.20
5	1.26	1.05	.52	-	-

[1,3] Calculated for Stokes numbers at S/W = 1, T/W = 1, and optimum inlet design conditions as defined by Marple[6]:
[1] Reynolds number at 0.21 m^3/min = 400
[3] " " " 1.13 m^3/min = 2000

[2] Solid particles; Whatman #41 oiled impaction surface.

[4] Liquid particles; glass fiber impaction surface, from Willeke[10]



NOTE: Slotted impaction substrate is shown in place on the collection plates.

Figure 1. Cross sectional diagram of the Sierra Model 235 slotted cascade impactor.

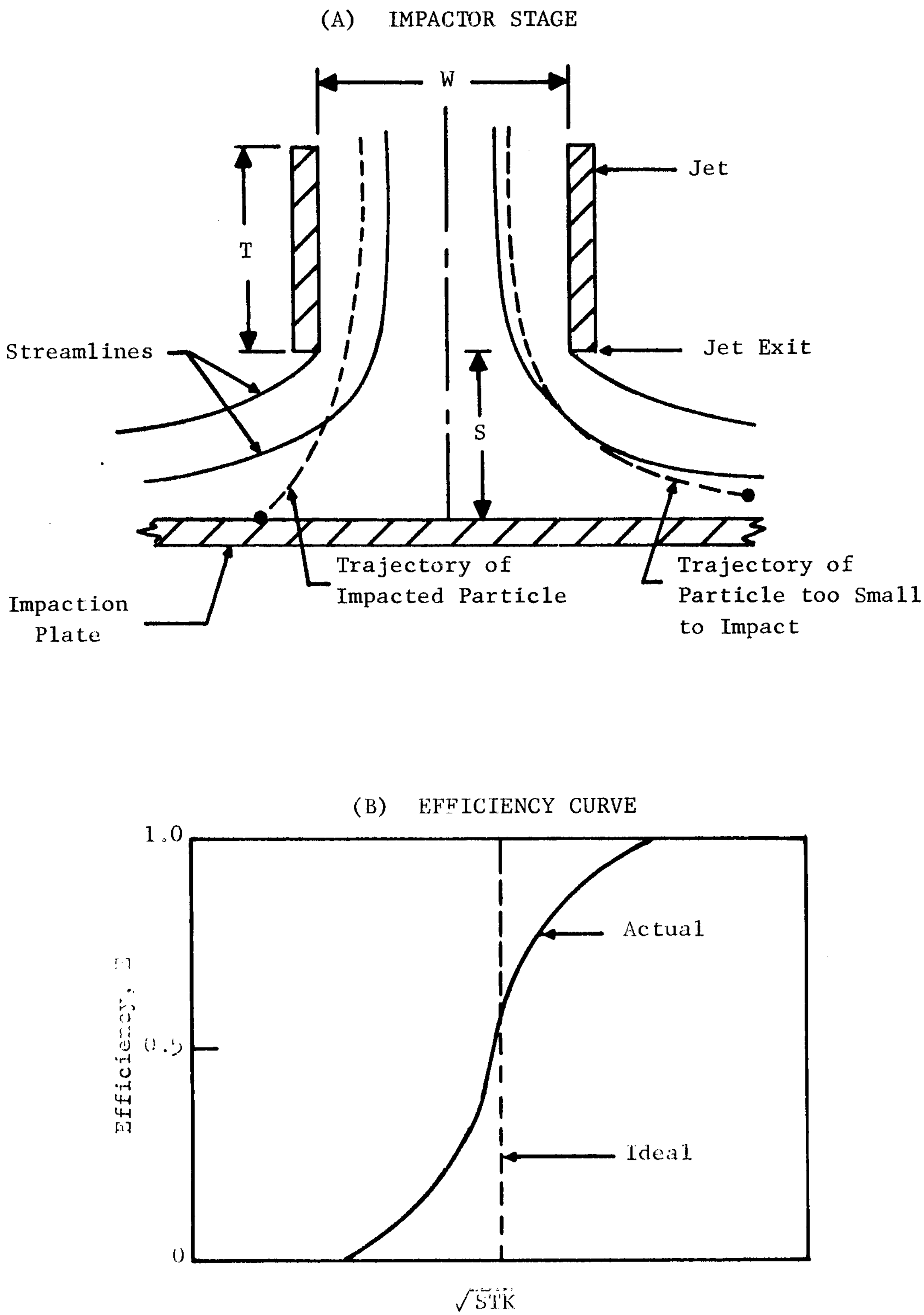


Figure 2. (A) Impactor flow lines and particle trajectories.
(B) Ideal and experimental efficiency curves (Marple[6]).

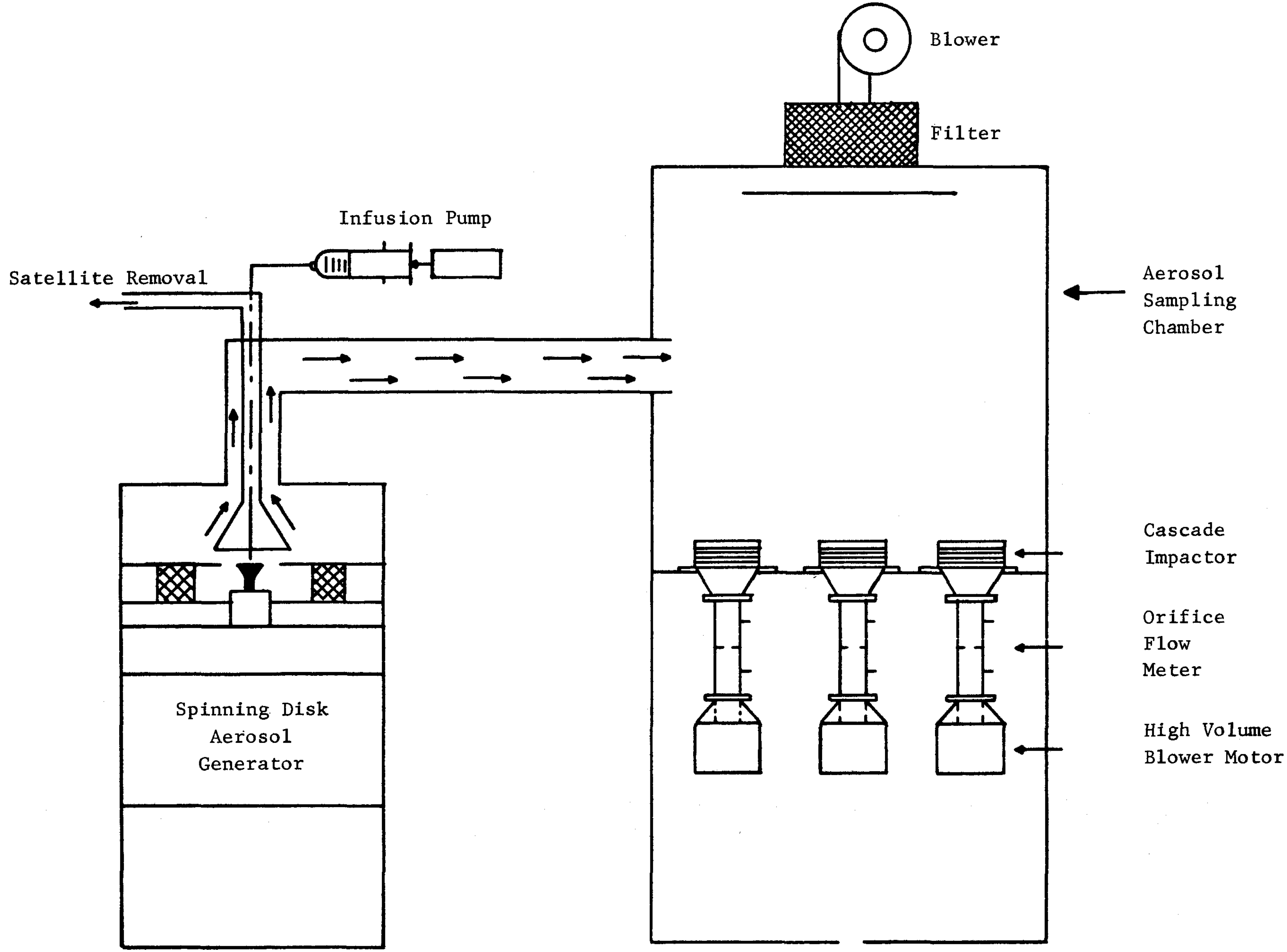


Figure 3. Cascade impactor calibration arrangement.

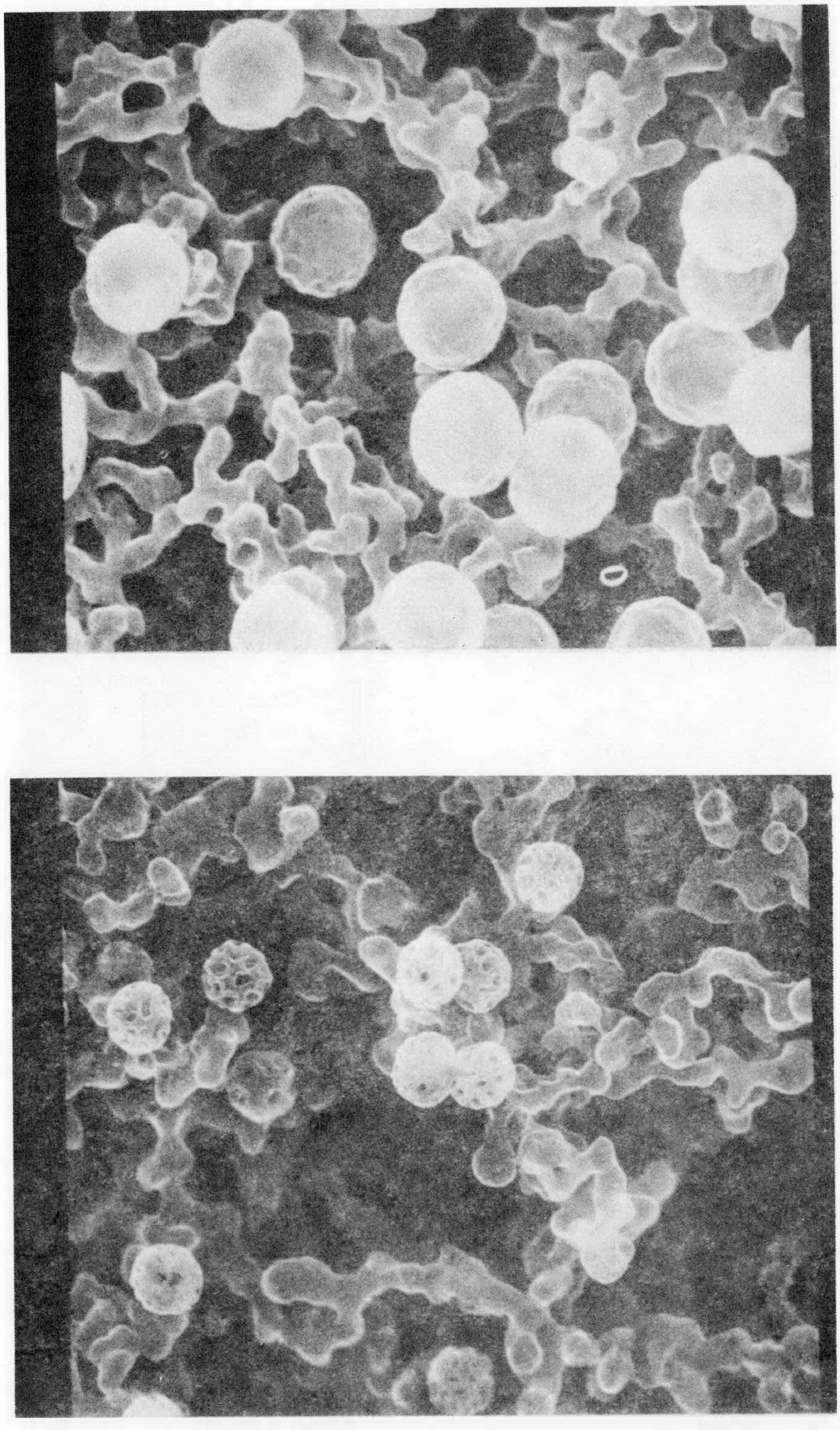


Figure 4. Scanning electron micrographs of test aerosol, < 2 μm, depicting sponge-like structure of particles.

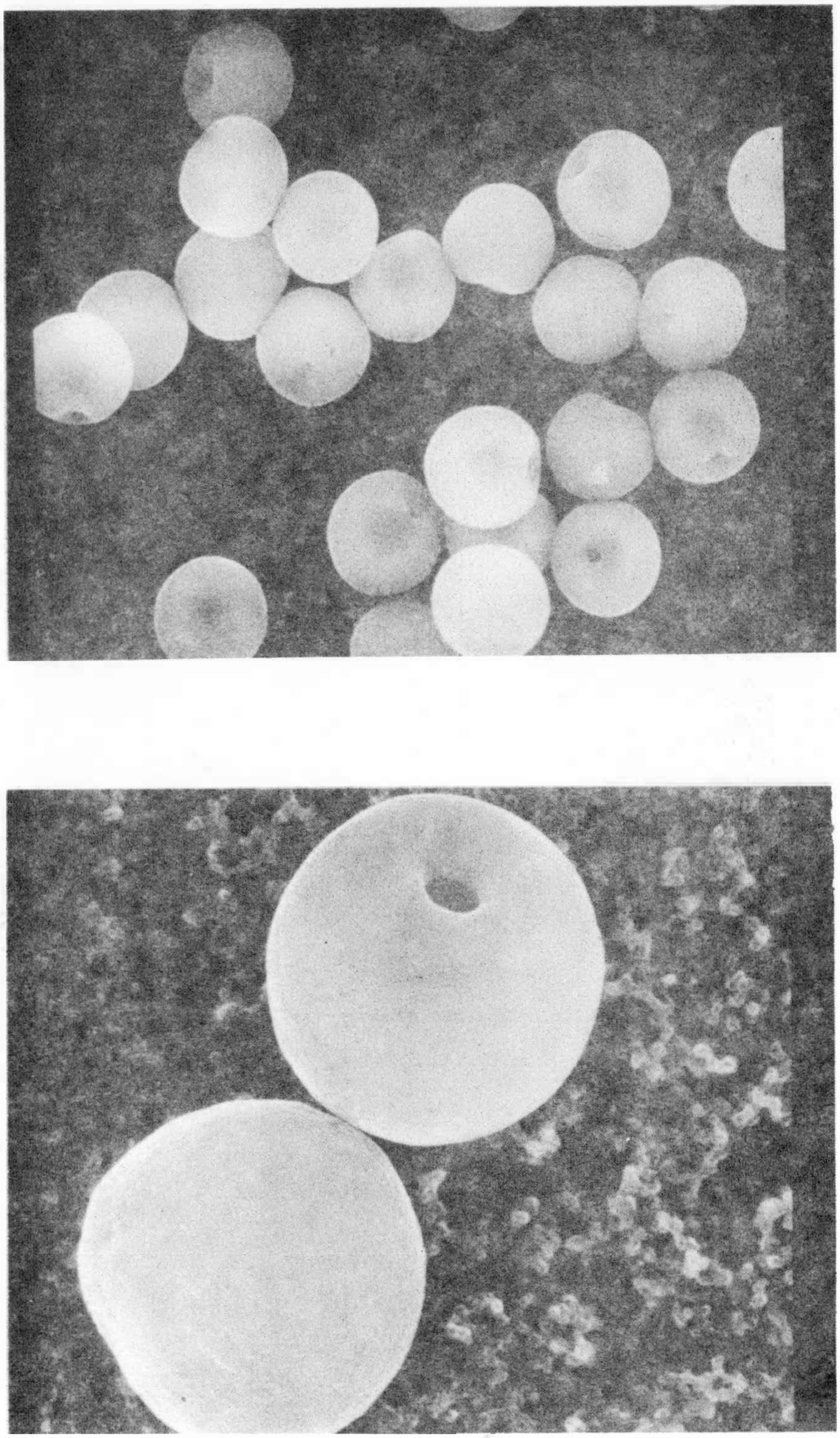


Figure 5. Scanning electron micrographs of test aerosol, > 15 μm, with internal voids.

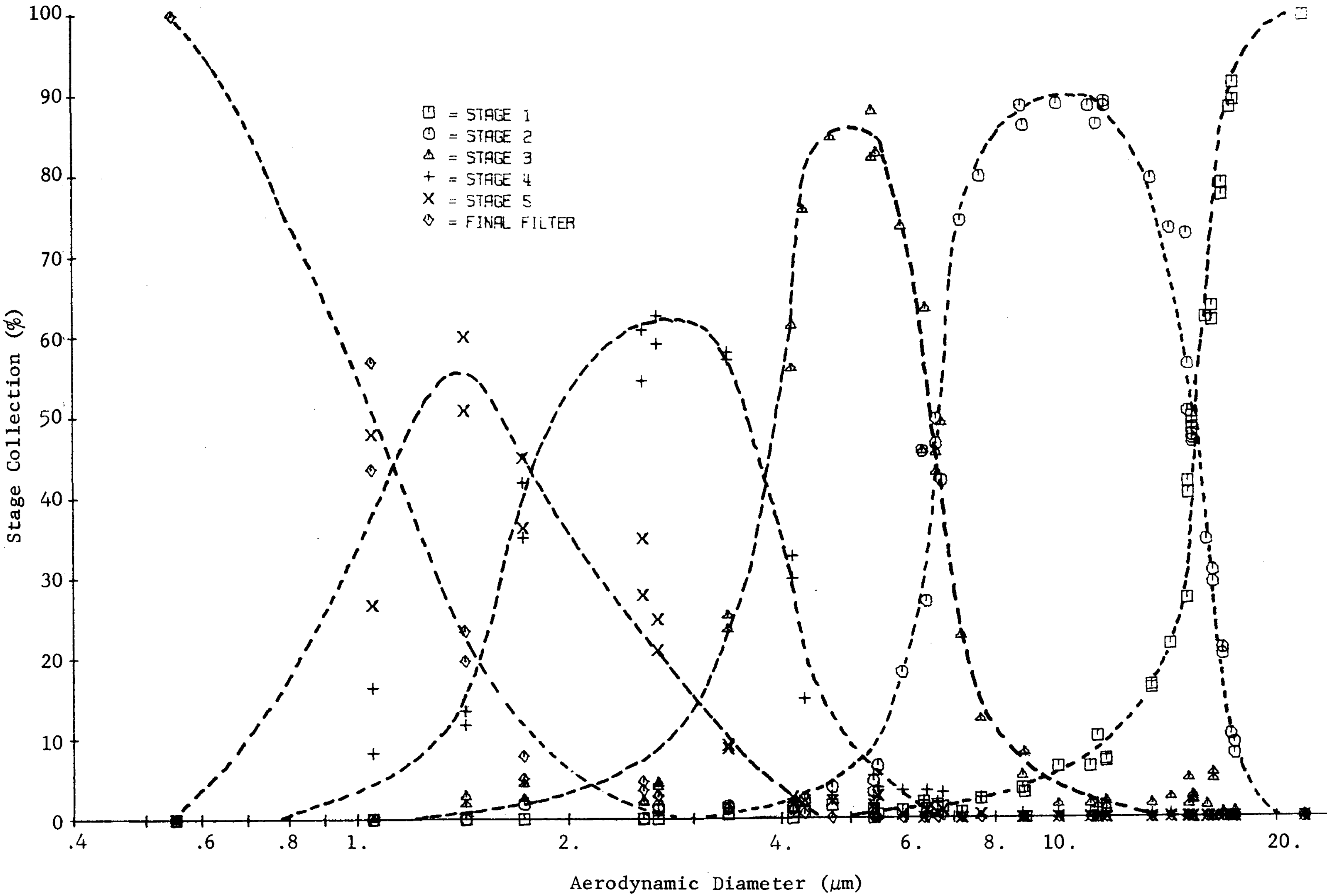


Figure 6. Stage collection curves for the Sierra Model 235 impactor at a flow rate of 0.21 m^3/min. Impaction surface is Whatman #41 with mineral oil.

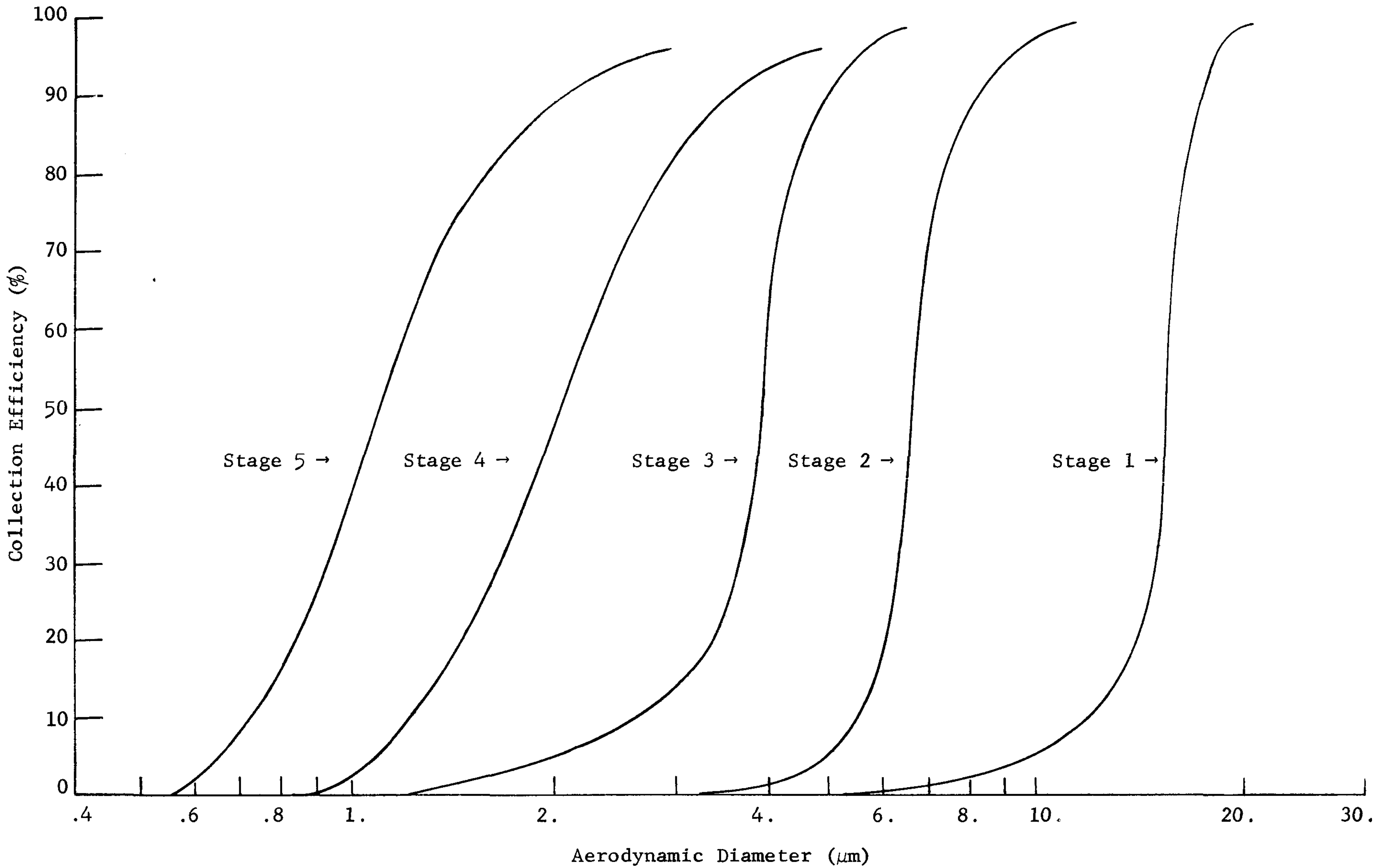


Figure 7. Efficiency curves developed from collection curves for the Sierra Model 235 impactor at a flow rate of 0.21 m^3/min. Impaction surface is Whatman #41 with mineral oil.

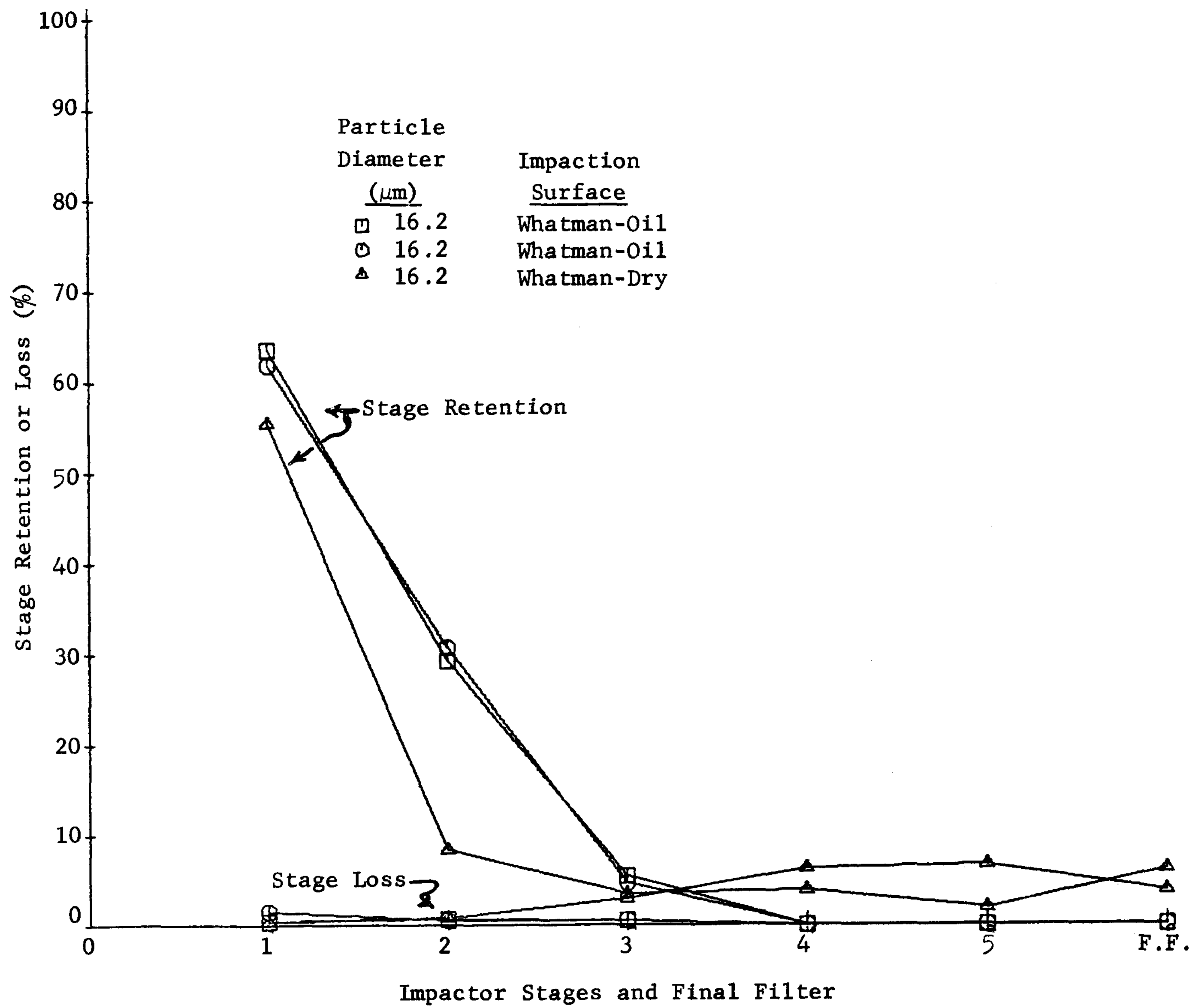


Figure 8. Effect of particle rebound on stage collection for the Sierra Model 235 impactor at a flow rate of 0.21 m^3/min.

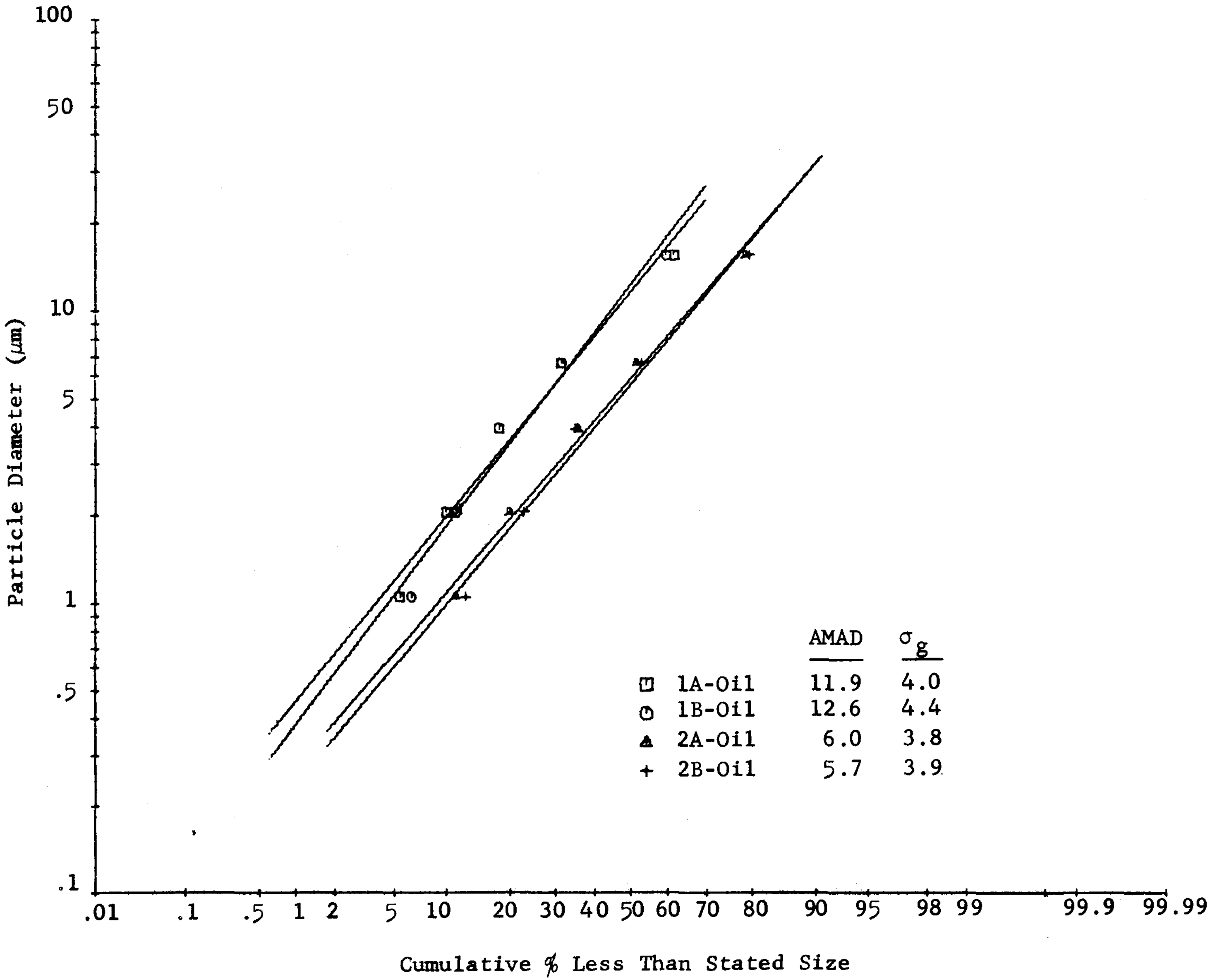


Figure 9. Log-probability plots of duplicate samples having Whatman #41 oiled impaction surfaces.

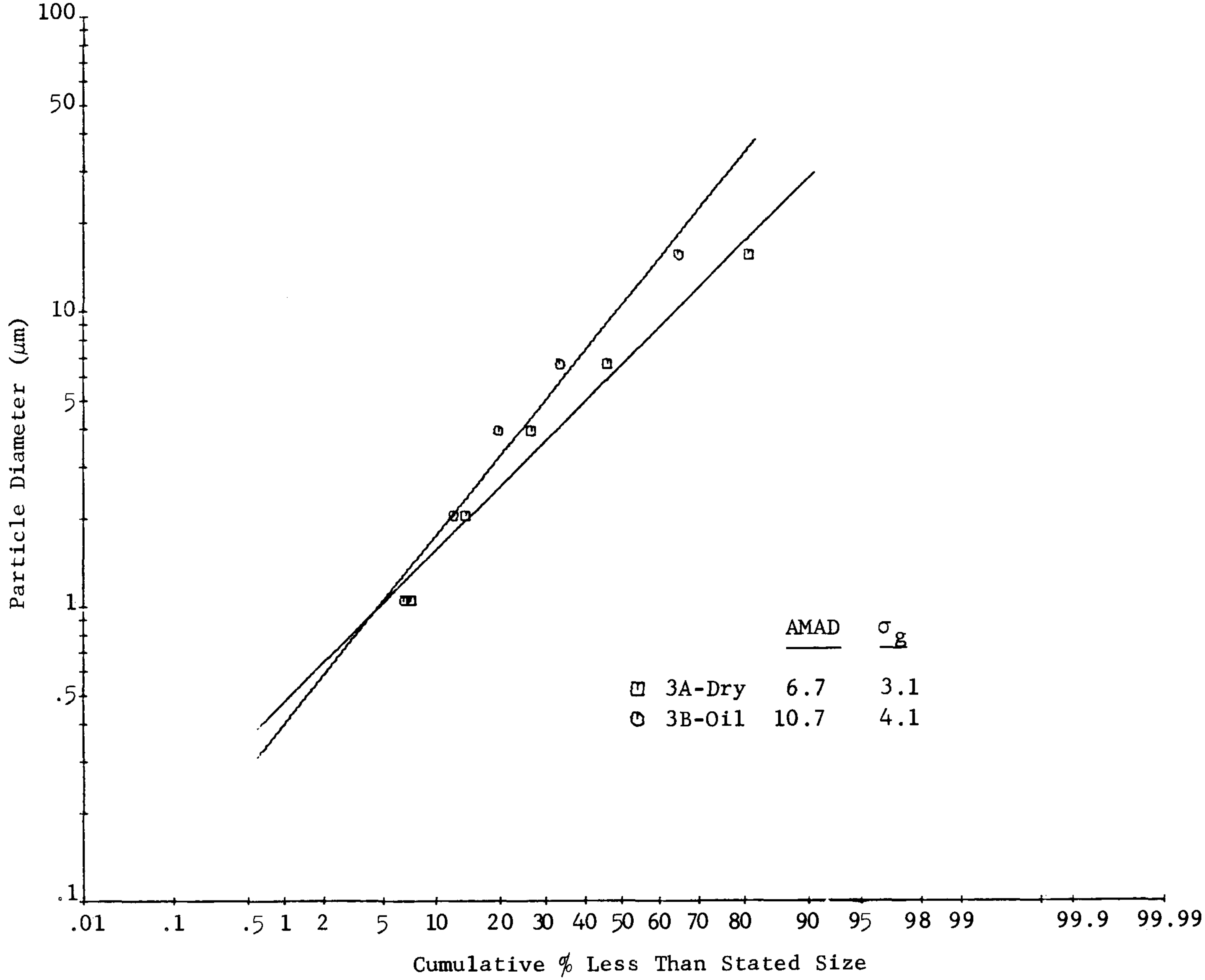


Figure 10. Log-probability plots of duplicate samples having respectively, dry and oiled Whatman #41 impaction surfaces.