

241
17-9-74
LA-6539-PR

Progress Report

UC-41

Issued: October 1976

Aerosol Research and Development Related to Health Hazard Analysis

July 1, 1975—June 30, 1976

Work performed by

Aerosol Studies Section
Industrial Hygiene Group

Lawrence W. Ortiz
Charles I. Fairchild
Marvin I. Tillery, Project Director
Harry J. Ettinger, Alternate Project Director

This work was supported by the National Institute for Occupational
Safety and Health, Project R072, Cincinnati, OH.



los alamos
scientific laboratory
of the University of California
LOS ALAMOS, NEW MEXICO 87545

An Affirmative Action/Equal Opportunity Employer

MASTER

UNITED STATES
ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION
CONTRACT W-7405-ENG. 36

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

The four most recent reports in this series, unclassified, are LA-5359-PR, LA-5555-PR, LA-5797-PR, and LA-6277-PR.

This work was supported by the National Institute for Occupational Safety and Health, Project R072, Cincinnati, OH.

Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, VA 22161
Price: Printed Copy \$3.50 Microfiche \$2.25

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights.

CONTENTS

ABSTRACT	1
I. FIBROUS AEROSOL STUDIES	1
II. FIBER SIZING	5
III. MISCELLANEOUS FIBER WORK	9
IV. FILTRATION EFFICIENCY OF GAS-VAPOR SAMPLER TUBES AGAINST PARTICULATES	9
V. REAL-TIME SAMPLER	15
VI. FUTURE WORK	16
REFERENCES	18

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

MASTER
iii

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

leg

AEROSOL RESEARCH AND DEVELOPMENT RELATED TO HEALTH HAZARD ANALYSIS

July 1, 1975 through June 30, 1976

by

Lawrence W. Ortiz and Charles I. Fairchild

Marvin I. Tillery, Project Director

Harry J. Ettinger, Alternate Project Director

ABSTRACT

Tests evaluating the British Mines Research Establishment (MRE) Horizontal Elutriator for sampling airborne fibrous glass were completed. Results confirm that impactor characterization of fibrous glass aerosols provides a reasonable approximation of the respirable mass fraction as defined by the American Conference of Governmental Industrial Hygienists (ACGIH) and the British Medical Research Council (BMRC) standards. Supplementary fiber-sizing data further indicate that the impactor provides diameter related size separation of fibrous glass aerosols. Test characterization of a new fibrous glass aerosol indicates that it is highly respirable material suitable for animal inhalation studies. Machine drawings of our fibrous aerosol generator have been made available to the public.

The particulate filtration efficiency of industrial gas-vapor sampler tubes was determined. This filtration efficiency is of interest for work environments containing toxic vapors and particulates of the contaminant, or inert particles containing adsorbed contaminant. The sampler tubes collect from 70-90% of the particles entering, but only 20% of the total is deposited on the active sorbent (charcoal granules). Considerable variability in collection efficiency between tubes was observed. Recommendations are made for obtaining improved contaminant estimates using these tubes.

The preliminary design of a real-time particle sizing sampler for use in coal mines is described.

I. FIBROUS AEROSOL STUDIES

Fiber sampling experiments evaluating the performance of the British MRE Gravimetric Dust Sampler (Horizontal Elutriator) with three different fibrous glass aerosols have been completed. These sampling experiments were conducted as part of the proposed sampler evaluation series intended to compare the sampling characteristics of a horizontal elutriator, 10-mm nylon cyclone, spiral centrifuge and Andersen impactor against various fibrous glass

aerosols. The MRE sampler (Fig. 1) is a portable two-stage sampler with a horizontal elutriator as the first stage followed by a total collection filter second stage. The aerodynamic separation characteristic of this instrument estimates the predicted lung deposition standard used by the BMRC¹ for compact (nonfibrous) particulates when operated at 2.5 l/min. All three fibrous aerosol materials tested with the MRE sampler were previously characterized by impactor sampling and were produced using conventional high-speed blenderization of bulk materials with subsequent generation by our modified Timbrell type generator.^{2,3} The first of these test aerosols was produced from Owens/Corning code AA-10 (O/C AA-10) material. Electron microscope (EM) sizing data indicate that the test aerosol produced from this material was composed of fibers with individual diameters ranging from ≈ 0.2 to ≈ 4.5 μm . MRE penetration results obtained against this O/C AA-10 fibrous glass aerosol indicate an average respirable mass of $49 \pm 3\%$. This average value is 7% higher than the 42% respirable mass previously obtained by Andersen impactor characterization of this same aerosol material⁴ [mass median aerodynamic diameter (mmad) = 4.0, geometric standard deviation (σ_g) = 1.7]. MRE results were the average of eight separate sampling runs conducted with the elutriator operating at the design flow rate of 2.5 l/min. Individual MRE penetration results obtained against this O/C AA-10 aerosol are summarized in Table I.

MRE sampling tests conducted against a second fibrous aerosol generated from another batch of Johns-Manville code 104 (J/M-104) fibrous glass indicate an average respirable mass fraction of $76 \pm 7\%$ for this test material. Andersen impactor characterization of this new J/M-104 test aerosol indicated a mmad of 2.7 μm with a σ_g of 1.6. These parameters define a respirable mass fraction of approximately 77% when correlated to the BMRC Lung Deposition Curve and a 64% respirable mass fraction when related to the ACGIH standard.⁵ These impactor recharacterization parameters (mmad = 2.7, σ_g = 1.6) are not significantly different from those values previously obtained against an earlier batch of J/M-104 material. (Previous impactor characterization of a J/M-104 aerosol produced a mmad = 2.9 with σ_g = 1.8, 59% respirable by ACGIH curve.) Impactor recharacterization tests were conducted against this new batch of material because original supplies of this same J/M-104 material were depleted prior to starting the MRE sampling series. Impactor recharacterization results are the average of seven separate impactor sampling runs conducted at random intervals during this MRE sampling series. Individual impactor recharacterization results are summarized in Table II. MRE results obtained against this new J/M-104 aerosol are the average of sixteen separate sampling runs conducted with the elutriator. Individual MRE penetration results are listed in Table III. Fiber-sizing results indicate that the test aerosol produced from this material was composed of predominantly individual fibers with a diameter range of ≈ 0.06 to ≈ 2.8 μm .

Similar MRE sampling tests conducted against a third aerosol produced from J/M-110 fibrous glass indicate an average respirable mass fraction of $31 \pm 4\%$ for this material. This approximates the $\approx 28\%$ respirable previously obtained by Andersen impactor characterization of a J/M-110 aerosol.⁶ MRE results are the average of sixteen separate sampling runs conducted with the elutriator. Individual MRE penetration results obtained against this test aerosol are summarized in Table IV.

Experimental MRE penetration data reported for all three fibrous glass aerosols tested were obtained during single 60-min MRE sampling runs. Elutriator penetration values were obtained by measuring the fibrous aerosol mass collected on a preweighed membrane filter located downstream of the four channel elutriator plates. The aerosol penetration fraction was determined by relating this mass fraction to the average challenge aerosol mass concentration measured by three separate gross filter samples collected simultaneously during four 15-min intervals for each 60-min sampling run. Thus, the average challenge aerosol mass was

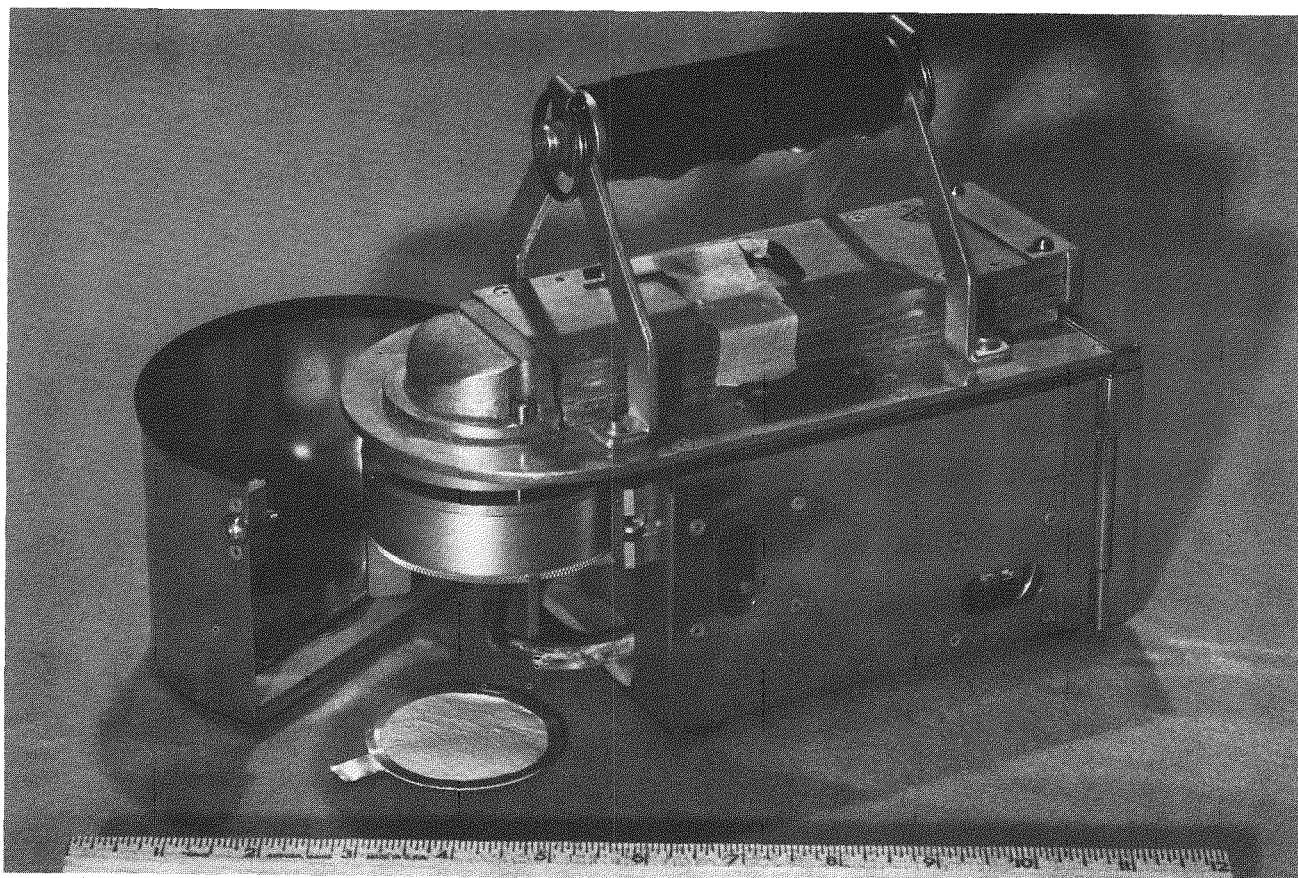


Fig. 1.
MRE Horizontal Elutriator, Type 113-A.

TABLE I
MRE HORIZONTAL ELUTRIATOR EVALUATION
O/C AA-10 FIBER GLASS^a

Reference Run Code	Elutriator Penetration (% wt)
J/M - 19342	
113	47
114	47
115	51
116	51
117	53
118	45
119	46
120	52
Average: 49	
Std Deviation: 3	
Coeff of Variation: 0.06	
No. of Tests: 8	

^aMRE Gravimetric Dust Sampler Type 113A
tested against blender-ground O/C AA-10
fibrous glass aerosol.

TABLE II
ANDERSEN IMPACTOR RECHARACTERIZATION OF
BLENDOR-GROUND J/M-104 FIBROUS GLASS AEROSOL

Run Code	Mass Median Aerodynamic Diameter (mmad- μ m)	Geometric Standard Deviation (σ_g)
J/M - 19558		
23	2.6	1.7
26	2.7	1.5
27	2.8	1.6
28	2.8	1.7
30	2.6	1.7
31	2.8	1.6
47	2.6	1.6
Average : 2.7		1.6
Std Deviation: 0.1		0.1
Coeff of Var: 0.04		0.05
No. of tests: 7		7

TABLE III

MRE HORIZONTAL ELUTRIATOR EVALUATION
J/M-104 FIBER GLASS^a

Reference Run Code	Elutriator Penetration (% wt)
J/M -19558-	
32	72
33	75
34	75
35	72
36	81
37	78
38	68
39	67
40	73
41	73
42	65
43	81
45	89
46	78
48	79
49	85

Average: 76

Std Deviation: 6.5

Coeff of Variation: 0.09

No. of Tests: 16

^aMRE Gravimetric Dust Sampler Type 113A
tested against blendor-ground J/M-104
fibrous glass aerosol.

TABLE IV

MRE HORIZONTAL ELUTRIATOR EVALUATION
J/M-110 FIBER GLASS^a

Reference Run Code	Elutriator Penetration (% wt)
J/M -19558-	
57	26
58	31
59	35
60	30
62	32
64	32
65	29
69	41
70	35
72	29
73	30
75	26
77	29
79	27
80	39
82	30

Average: 31

Std Deviation: 4

Coeff of Variation: 0.13

No. of Tests: 16

^aMRE Gravimetric Dust Sampler Type 113A
tested against blendor-ground J/M-110
fibrous glass aerosol.

determined by twelve separate open-face filter samples for each elutriator sampling run. Sampling flow through the elutriator was measured and controlled at 2.5 l/min using a calibrated rotameter attached to an externally operated carbon vane pump. Elutriator total collection backup filters used for gravimetric assessment were 55-mm-diam, Gelman VM-1, polyvinylchloride (PVC), 5.0- μ m pore, membrane filters. Open-face filter samples were collected on 25-mm-diam, Gelman DM-800 filters. Mass measurements obtained were well within the sensitivity and precision of the Mettler AD-1 microbalance used (± 5 μ gm).

These MRE sampling experiments complete the gravimetric evaluation portion of the horizontal elutriator test series against coarse J/M-110, medium O/C AA-10, and fine J/M-104 fibrous glass aerosols. Supplementary fiber-sizing experiments are currently scheduled for one of these same aerosols (J/M-110) to further provide a size/mass relationship for horizontal elutriator sampled fibrous glass aerosols. Specifically, these data will relate, by fiber size, the aerosol fraction penetrating the elutriator to the previously obtained challenge aerosol size distribution. Samples for sizing have been taken, and a nine-sample composite will be prepared for sizing by electron microscopy.

Table V summarizes individual results obtained against the three different fibrous glass aerosols tested with the MRE sampler. In each case, aerodynamic characterization was determined using the Andersen impactor, and fiber diameter measurements were obtained from sizing collected aerosols by electron microscopy using our membrane filter to EM grid transfer procedure.⁶ Impactor characterization was based on a gravimetric assessment of the cumulative fibrous aerosol mass collected on successive impaction stages with m_{mad} and σ_g

TABLE V
SUMMARY OF IMPACTOR/ACGIH/BMRC/MRE CORRELATIONS AGAINST
THREE SEPARATE FIBROUS GLASS TEST AEROSOLS

Test Fibrous Aerosol Designation	Aerosol Size Range by Individual Fiber Diameter (μm)	Impactor Mass Characterization ($\text{mmad}-\mu\text{m}$) (σ_g)		Predicted "Respirable" Fraction From Impactor Characterization		Average MRE ^a Penetration Values (%)
				ACGIH Curve (%)	BMRC Curve (%)	
J/M-110	≈ 0.07 to ≈ 7.0	5.1-6.1	1.6-2.0	≈ 28	≈ 39	≈ 31
O/C AA-10	≈ 0.2 to ≈ 4.5	4.0	1.7	≈ 42	≈ 57	≈ 49
J/M-104	≈ 0.06 to ≈ 2.8	2.7	1.6	≈ 64	≈ 77	≈ 76

^aMRE is designed to satisfy the BMRC definition of respirable dust.

determined from a log-probability plot related to this experimentally measured cumulative mass distribution. The mmad and σ_g were used to calculate the respirable mass fraction of these aerosols for the ACGIH and the BMRC Respirable Dust Curves. MRE results are the average values experimentally obtained by the procedure discussed above. Comparison of these summary results indicates that impactor mass characterization of fibrous glass aerosols provides a reasonable approximation of the respirable mass fraction as defined by either respirable dust curve.

II. FIBER SIZING

Fiber-sizing data have been obtained from Andersen impactor samples of an O/C AA-10 fibrous glass aerosol. These data can be used to relate the size distribution by individual fiber diameter of fibrous material collected on successive impaction stages to aerodynamic size as defined by the impactor. Sizing data were obtained from composite samples prepared for each successive impaction stage from five separate impactor sampling runs. All samples for sizing were collected on membrane filters, composited by rinsing each impaction stage collection filter into separate corresponding reservoirs of toluene, dispersing composite suspensions with ultrasonic agitation, filtering aliquots of each composite dispersion onto clean membrane filters, and sizing individual collected fibers by electron micrographs taken with our membrane filter to EM grid transfer procedure.⁶ Table VI summarizes the average fiber diameters and the range of fiber diameters measured for each impaction stage as related to the corresponding Andersen impactor effective cutoff diameter (ECD) for that same stage. These data indicate a trend of fibers being collected in an order of progressively decreasing diameters on successive impaction stages. Impactor collection stages numbered 0 and 1 (Andersen ECD: 11 and 7 μm) produced an average measured fiber diameter of 1.2 μm and 1.5 μm , respectively. This arrangement of average size order for these two stages is not unreasonable as aerosol material collected on these first two stages reflects the actual composition of the challenge aerosol. That is, the aerosol fraction collected on stages 0 and 1 contains material that existed, in the airborne state, as a composite of fibrous flocks, loosely bound fiber agglomerates as well as large fiber singlets. These airborne fibrous flocks and agglomerates can be composed of various combinations of small-, medium-, and large-diameter fibers which would be collected as aerodynamically large particles. The corresponding measured size data for all stages reflect, in contrast, collected fibers as sized only according to individual fiber diameter. The phenomenon of aerodynamic size fractionation as related to fiber diameter, becomes apparent beginning with impaction stage 2. This is to be expected as the probability of fiber bundles penetrating each successive impaction stage decreases in the same order. The size order for fibers collected on subsequent impaction stages (by average fiber diameter) then proceeds: 1.2 μm for stage 2, 1.1 μm for stage 3, 0.7

μm for stage 4, $0.5\ \mu\text{m}$ for stage 5, and $0.3\ \mu\text{m}$ for the backup filter. These results are listed in Table VI. The inclusion of appropriate shape factors and density to the average fiber diameter results in good agreement between average fiber diameter and effective cutoff diameter for stages 2 through 5, thus indicating the prime importance of fiber diameter in determining the aerodynamic characteristics of fiber particles.

Figures 2 through 8 illustrate the respective fiber number distributions, by individual fiber diameter, obtained for this O/C AA-10 aerosol as collected on each successive impaction stage. Each successive frequency distribution histogram relates aerodynamic size information (Andersen ECD) for a single impactor stage, to the distribution of sampled fiber diameter. Examination of each distribution histogram in sequence confirms the trend of aerosol fractionation according to sampled fiber diameter. This trend, as mentioned above, is not readily apparent until the aerosol reaches impaction stage 2. At stage 2 only 0.5% of the individual fibers measured were $\geq 3.1\text{-}\mu\text{m}$ diam while in comparison stages 0 and 1 had 5.0% and 6.5% $\geq 3.1\text{-}\mu\text{m}$ diam, respectively. These data indicated that the large diameter ($\geq 3.1\ \mu\text{m}$) fibers had been removed from the challenge dust by the time the aerosol reached impaction stage 2. The aerosol fractionation trend becomes more pronounced with successive collection stages: Stage 3 had no fibers $> 3.0\text{-}\mu\text{m}$ diam with $\approx 83\%$ of measured fibers being $< 1.6\text{-}\mu\text{m}$ diam; stage 4 had no fibers $\geq 2.1\text{-}\mu\text{m}$ diam with $\approx 93\%$ $\leq 1.1\text{-}\mu\text{m}$ diam; stage 5 had no fibers $> 1.2\text{-}\mu\text{m}$ diam with $\approx 93\%$ $\leq 0.7\ \mu\text{m}$ and the backup total-collection filter had only 11% of the measured fibers $> 0.5\ \mu\text{m}$ with $\approx 89\%$ $\leq 0.4\text{-}\mu\text{m}$ diam. These sizing data are the result of measuring > 4300 individual impactor collected fibers. Both fiber diameter and fiber length were measured when possible, but in most cases individual fibers were too long for proper sizing by electron microscopy. (This applied to fibers collected on all six impactor collection stages.)

This observed mode of impactor fractionation/collection as applied to this fibrous glass aerosol further confirms the contention that the aerodynamic behavior of airborne fibers is largely diameter related.^{2,4,7,8} Furthermore, these impactor sizing data also suggest that impactor mass characterization provides size (fiber diameter) related aerodynamic information for sampled fibrous glass aerosols.

The phenomenon of fiber fractionation by impactor also suggests a practical technique for the size selection of fibers which should be of use to investigators interested in obtaining quantities of discretely sized fibers. Proper choice of starting materials coupled with selected cascade impactor fractions could rapidly provide gram quantities of fibers in narrow diameter ranges. Fibers fractionated according to diameter are currently of special interest for use in animal inhalation/implantation experiments.

A sample of new fibrous glass material was milled, generated, and characterized as part of our continuing fiber sampling evaluation program. This new fibrous glass material (J/M-100) is composed of extremely fine diameter fibers. (Manufacturers claim $\approx 0.05\text{-}\mu\text{m}$ average fiber diameter). This material was easy to mill, and sampling studies indicate that the aerosol produced has a large fraction of material in the respirable range. Andersen impactor characterization of this test aerosol produced a mmad of $2.6 \pm 0.1\ \mu\text{m}$ with $\sigma_g = 1.9 \pm 0.1$. These results are the average of four separate impactor screening runs. Individual impactor results are listed in Table VII. These impactor measured parameters (mmad = $2.6\ \mu\text{m}$, $\sigma_g = 1.9$) define a respirable mass fraction of approximately 74% based on the BMRC Lung Deposition Curve and a 62% respirable mass fraction based on the ACGIH standard. MRE Horizontal Elutriator screening results obtained against this same aerosol produced an $81 \pm 3\%$ mass fraction penetrating the elutriator. This is the average of three separate elutriator runs. Individual MRE sampling results are listed in Table VIII. These impactor/MRE sampling results indicate that this J/M-100 material may be well-suited for animal inhalation studies and again confirm use of impactor sizing data to predict respirable fraction. Cyclone calibration tests are not scheduled for this aerosol.

TABLE VI
ANDERSEN IMPACTOR SIZE CHARACTERIZATION OF
O/C AA-10 FIBROUS GLASS AEROSOL

Impactor Parameters		Collected Fiber Characteristics		
Stage	Assigned Effective Cutoff Diameters	Fiber Diameter Range	Average Fiber Diameter	No. of Fibers
No.	FCDs - (μm)	(μm)	(μm)	Sized per Stage
0	11	0.1 to 5.3	1.2	325
1	7	0.5 to 5.5	1.5	544
2	4.7	0.3 to 4.0	1.2	621
3	3.3	0.2 to 2.9	1.1	524
4	2.1	0.1 to 2.0	0.7	790
5	1.1	0.2 to 1.6	0.5	807
Backup	<1.1	0.1 to 1.5	0.3	729

Total No. of Fibers Sized: 4340

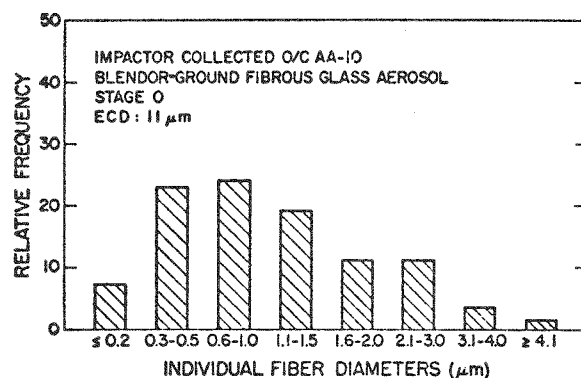


Fig. 2.
Fiber number distribution by diameter.

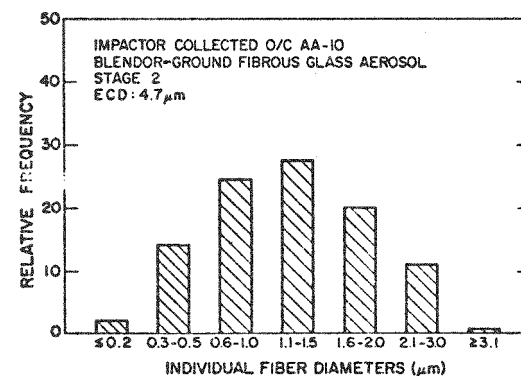


Fig. 4.
Fiber number distribution by diameter.

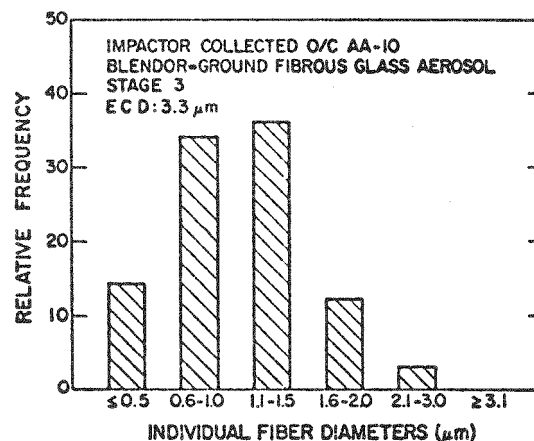


Fig. 5.
Fiber number distribution by diameter.

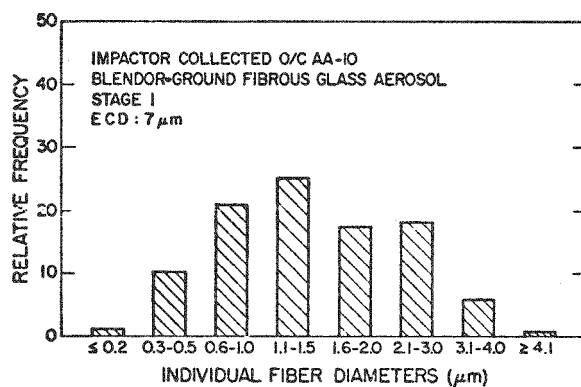


Fig. 3.
Fiber number distribution by diameter.

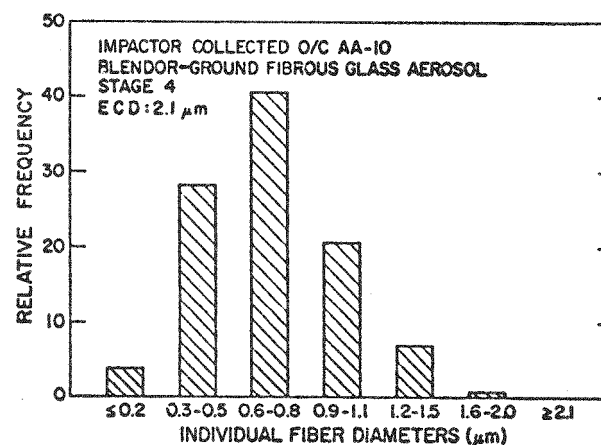


Fig. 6.
Fiber number distribution by diameter.

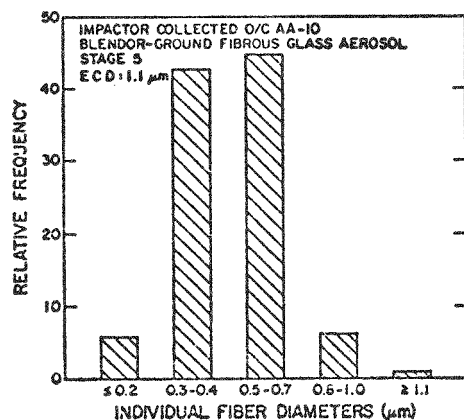


Fig. 7.
Fiber number distribution by diameter.

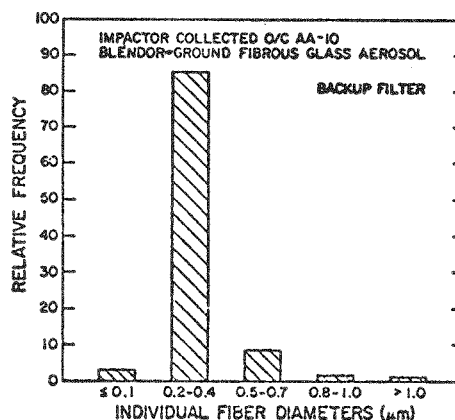


Fig. 8.
Fiber number distribution by diameter.

TABLE VII
ANDERSEN IMPACTOR CHARACTERIZATION OF
BLENDOR-GROUND J/M - 100 FIBROUS GLASS AEROSOL

Reference Run Code	Mass Median Aerodynamic Diameter ($m_{mad} - \mu m$)	Geometric Standard Deviation (σ_g)
J/M -19558-		
83	2.6	1.9
84	2.6	1.8
85	2.7	1.9
86	2.5	1.8
Average:	2.6	1.9
Std Deviation:	0.1	0.1

TABLE VIII
MRE HORIZONTAL ELUTRIATOR EVALUATION OF BLENDOR-
GROUND J/M - 100 FIBROUS GLASS AEROSOL

Reference Run Code	Elutriator Penetration (% wt)
J/M -19558	
89	79
90	84
91	80
Average:	81
Std Deviation:	3

Fiber-sizing data have been obtained against an aerosol produced from blendorized Gelman Type A fibrous glass. These data indicate that the gross aerosol produced from this material contains fibers with individual diameters ranging from 0.07 to 3.0 μm . The average fiber diameter resulting from 426 individual measurements for this aerosol was $\approx 0.5 \mu m$. All samples for sizing were collected on membrane filters and sized by electron micrographs taken using our membrane filter to EM grid transfer procedure. These sizing experiments complete the gross aerosol measurement sequence for the seven different fibrous glass aerosols originally tested against the 10-mm nylon cyclone. These data provide supporting evidence that the experimentally obtained values of the aerodynamic parameters m_{mad} and σ_g of this particular aerosol are reasonable as related to individual fiber diameter measurements comprising the same test aerosol. Previous impactor characterization of this Gelman Type A aerosol produced a m_{mad} of 3.1 μm with a σ_g of 1.7.⁴ Similar data obtained against other test fibrous glass aerosols indicate that these aerodynamic characterization values are reasonable for a fibrous glass aerosol having an average individual fiber diameter of $\sim 0.5 \mu m$.

III. MISCELLANEOUS FIBER WORK

Reports

A topical report, describing our existing fibrous aerosol generator is currently being prepared. This report contains information on materials, construction, design, and performance of our existing generator. Detailed machine drawings of this generator were prepared and copies sent to NIOSH, Cincinnati, Ohio, and to the Technical Information Center, Oak Ridge, Tennessee. These drawings involved a major effort and consist of twelve separate 24-in. by 36-in. sheets containing sixty-plus separate drawings detailing components and construction of the generator assembly. Copies of these drawings are available through the Technical Information Center, U.S. ERDA, Box 62, Oak Ridge, Tennessee, 37830, Reference Drawing Case No. 2429, Fibrous Aerosol Generator. These drawings should be of value to other investigators who desire a reliable mechanism for generating fibrous aerosols.

Spiral centrifuge characterization of a single fibrous glass aerosol (J/M-104) will complete our intersampler comparison series as originally proposed. Sampling runs of 20-, 20-, 40-, and 100-h duration have been made and collection grids are currently being sized. EM grids placed at 10-cm intervals along the 180-cm spiral collection foil should provide aerodynamic information for the spectrum of test aerosol deposited along the foil length. Aerodynamic characterization for this portion of the intersampler comparison series will be by equivalent size correlation only (not mass). EM sized fibers deposited on the collection foil at specified intervals will be related to the corresponding deposition of aerodynamically established standards such as unit density polystyrene latex (PSL) spheres.

NIOSH Proficiency Analytical Testing (PAT) asbestos loaded membrane filters are being counted on a regular basis. The microscopical appearance of the last seven sets (NIOSH PAT Rounds 33-39) have been excellent.

IV. FILTRATION EFFICIENCY OF GAS-VAPOR SAMPLER TUBES AGAINST PARTICULATES

Air-sampling tubes filled with a sorbent material are commonly used in industry to monitor personal exposure to toxic vapors and gases. Although these sampling tubes are designed to sample gases, they are frequently used in environments that contain gases, vapors, and particulates. Because airborne particles may contain adsorbed contaminant or be composed of volatile solid or liquid contaminant, the collection of such particles by the gas sampling tube is important.

Commercial tubes used for sampling organic vapors were studied. These 4-mm-diam glass tubes contain two sections of 20/40 mesh activated charcoal granules as the sorbent. The sorbent sections are held in place and separated by one glass wool and two polyurethane retainer plugs. An entire personal sampling system is shown in Fig. 9 with detail of the gas-vapor sampler (G-V-S) shown in Fig. 10. All components of the G-V-S will collect some fraction of the particles that enter the tube. However, only the sorbent material is analyzed for contaminant; the other components are discarded, possibly with a significant portion of the contaminant. Consequently, the particle collection efficiency of each component may be important to the sample analysis.

Filtration efficiencies were determined against four monodisperse and one polydisperse aerosol. The aerosols were 0.3- μ m dioctyl phthalate (DOP), 0.8-, 1.0-, and 2.0- μ m polystyrene latex (PSL), and polydisperse PSL with a count median diameter (CMD) of 5.7 μ m and a σ_g of 1.4. A Model Q-127 thermal generator⁹ provided the DOP aerosol, and the PSL

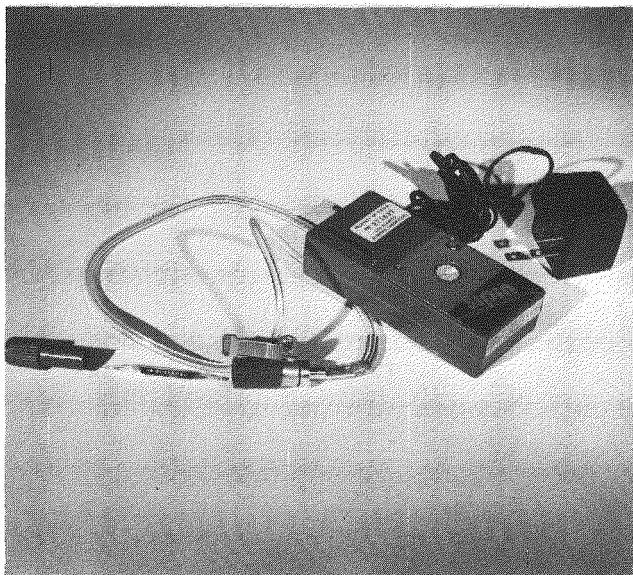


Fig. 9.

Organic vapor personal sampler unit and recharger.

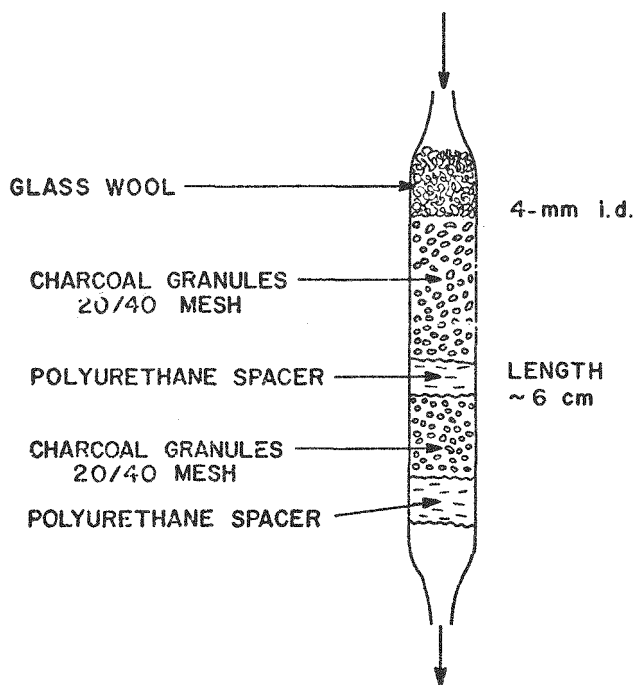


Fig. 10.

Organic vapor sampler tube.

aerosols were generated with RETEC medical nebulizers¹⁰ operated at 7.5 psig. The experimental system is shown in Fig. 11. The solution in the nebulizer was 1 part of Dow 10% PSL solution and 450 parts of double distilled water. The aerosol is first introduced into the drying chamber and mixed with approximately 20 l/min of clean dilution air. The aerosol was passed through a tritium electrical charge neutralizer to reduce particle charge. The challenge aerosol then passed into a small chamber located just upstream of the G-V-S tube. Samples were collected from this chamber on preweighed membrane filters. The aerosol concentration was also measured using a forward light-scattering photometer. A HEPA filter rated at 850 l/min with a pressure drop of 2.0 cm of water was connected to the chamber to remove excess aerosol and insure the inlet pressure of the G-V-S tube was approximately ambient. The penetrating concentration was also measured by a light-scattering photometer and/or other instrumentation. For a few tests conducted at a flow of 200 cm³/min the challenge aerosol was drawn through the G-V-S tube and the downstream measurement system by the Sipin pump shown in Fig. 9. In the remaining tests, to minimize sedimentation losses due to low flow velocities through the G-V-S tube and downstream detector, clean supplemental air was added between the G-V-S tube and downstream detector through a flow balancing dilution system (Fig. 11).

Gravimetrically determined challenge concentrations ranged from 1 to 5 µg/l for all PSL aerosols except the 5.7-µm CMD PSL. The DOP aerosol concentration ranged from 7 to 15 µg/l. Nebulization problems and transport line losses limited the challenge concentration of the 5.7-µm CMD PSL aerosol to less than 1 µg/l. Samples for examination by electron

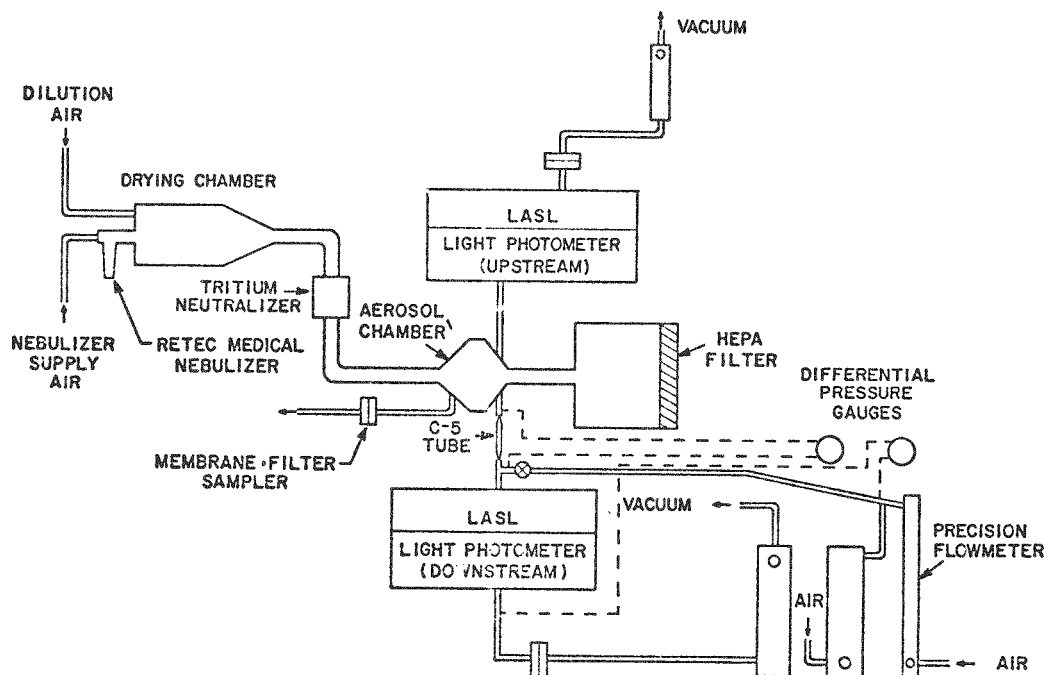


Fig. 11.
Apparatus to measure particulate filtration efficiency.

microscopy were collected with an electrostatic precipitator to determine the number of multiple particles in the challenge aerosol. Filtration efficiency measurements were not made if more than 10% of the challenge aerosol consisted of multiple particles.

Penetration efficiencies were measured for flows of 50, 100, and 200 cm³/min, which translate to average velocities of 7, 13, and 27 cm/s through the 4-mm-i.d. tubes. For monodisperse aerosols, penetration measurements were made using two forward light-scattering photometers calibrated against the challenge aerosol concentration before and after each penetration test. Penetration was determined by monitoring the upstream and downstream aerosol concentration. The photometers were designed and constructed in house and have a sensitivity for 0.3- μ m DOP of 15 particles/cm³.¹¹ A mass measuring system provided better accuracy for the 5.7- μ m polydisperse PSL aerosol. For this aerosol, upstream mass concentration was measured gravimetrically with membrane filter samples and downstream concentration was measured by a piezoelectric crystal particle mass monitor (PMM).¹²

The precision of the light-scattering photometer method was determined to be $\pm 7.5\%$ of full scale whereas that of the PMM was $\pm 3.5\%$ of full scale. Since the aerosol penetration changed with time the precision could not be measured under constant conditions. Consequently, precision is defined as the square root of the sum of the squares of the data deviations from a linear correlation of the data. The precision was determined in this way for each run and then averaged to obtain the above figures.

The relative accuracy of each instrument was determined by comparing measurements by photometers and the PMM to membrane filter measurements, using PSL aerosols at different concentrations. Figure 12 illustrates the relative accuracy of the photometers and the PMM. The horizontal line at 1.0 represents agreement with the concentration obtained gravimetrically and the shaded area indicates the relative error band expected from weighed samples obtained from a fixed volume of 500 l of aerosol at the indicated concentration. The

ratio of the concentration indicated by the PMM or light photometers to that indicated gravimetrically (relative accuracy) is plotted for each instrument as a function of aerosol concentration.

The relative accuracy of the light photometers was better than the PMM at low concentrations for the small particle aerosols; whereas, the accuracy of the PMM was superior to the photometers for the 5.7- μm CMD aerosol at all concentrations. The relative accuracy of both systems decreases at low concentrations (Fig. 12). Thus, the upstream concentration was made as high as practicable so that the downstream or penetrating aerosol concentration would also be relatively high. Of course, the gravimetric method would be the most accurate at low aerosol concentration, but the time required to collect sufficient downstream sample becomes prohibitive.

Overall penetration of sampling tubes as a function of time is shown in Figs. 13-15 for three flow rates and five different test aerosols. The greatest penetration occurred for 0.8-1.0- μm PSL aerosol, whereas the 5.7- μm polydisperse PSL exhibited the lowest penetration. During these tests a decay in penetration was observed over the sampling period when the initial penetration was greater than 15%. When the initial penetration was less than 15% the value stayed essentially constant over the 120-min sampling period. These results were unexpected as it would seem that the more efficient collectors would show greater loading effects. A wide variability in the rate of penetration decrease and shape of the penetration decrease curve was noted. In some cases penetration decreased rapidly then remained essentially constant for the rest of the run. In other cases there was an essentially constant decay in penetration over the 120-min sampling period. For lack of a better model, penetration was assumed to decay at a constant rate if initial penetration was $>15\%$, or

$$P(t) = P_i - At \quad (1)$$

where $P(t)$ is the penetration at time $t(\text{min})$, P_i initial penetration, and A a constant. A was found to equal 0.03 ± 0.03 when averaged over all particle sizes. Individual run data produced linear correlation coefficients (r , Figs. 13-15) ranging from 0.3 to 0.9 with an average value of $0.6 \pm .2$.

The results from these tests also verified that penetration varies greatly from tube to tube, with the mean reproducibility from tube to tube being $\pm 55\%$ (coefficient of variation for 71 tubes tested). This variation could be expected since these tubes were not designed to collect particulates.

Experiments were conducted to determine the aerosol penetration through each component in the sampling tube. Each G-V-S tube contained loosely packed glass wool in the inlet (Fig. 10), followed by a 15-mm-long section of charcoal, a polyurethane spacer, a 7.5-mm-long section of charcoal, and finally another polyurethane spacer. These tests used only the 1.0- μm -diam monodisperse PSL aerosol at a single flow rate of 200 cm^3/min . The procedure was: (1) obtain an overall G-V-S sampler penetration during 100 min of sampling, (2) remove the glass wool inlet plug and obtain penetration without this component, (3) remove the charcoal to obtain the penetration of the polyurethane spacers only, and (4) remove the plugs to obtain the empty tube and plumbing penetration efficiency. Penetration of the charcoal alone could not be obtained directly because without support the loose charcoal granules would not remain in a coherent body. Each of five G-V-S tubes was tested in this way. The averaged results are graphed in Fig. 16.

From these penetration results the collection efficiency of each section, including the charcoal, can be calculated. The material lost on the tube walls is small ($<6\%$) and will be ignored. The collection efficiency of the two polyurethane spacers is calculated first since these

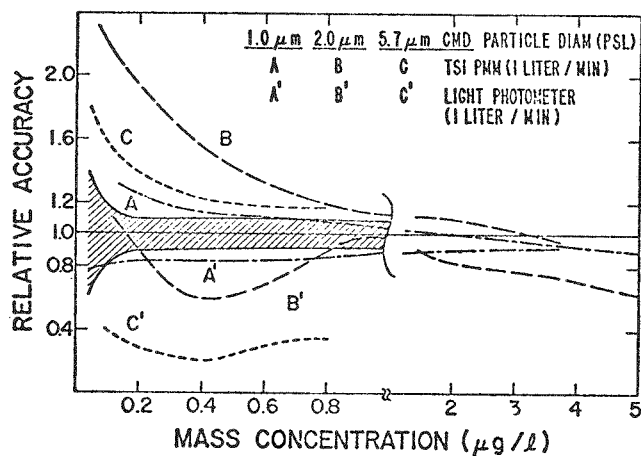


Fig. 12.
Light-scattering photometer and particle mass monitor accuracy relative to gravimetric measurement.

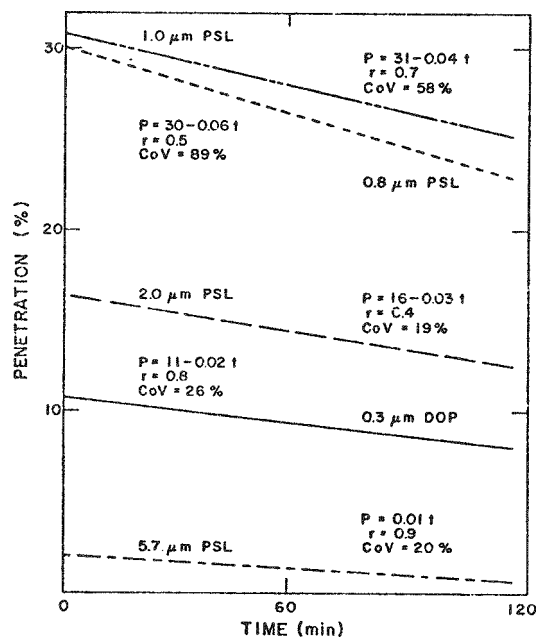


Fig. 13.
Aerosol penetration through G-V-S tubes at 50 cm^3/min .

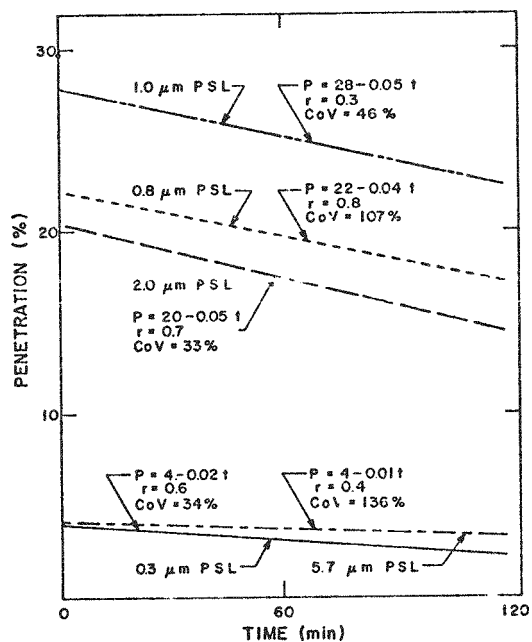


Fig. 14.
Aerosol penetration through G-V-S tubes at 100 cm^3/min .

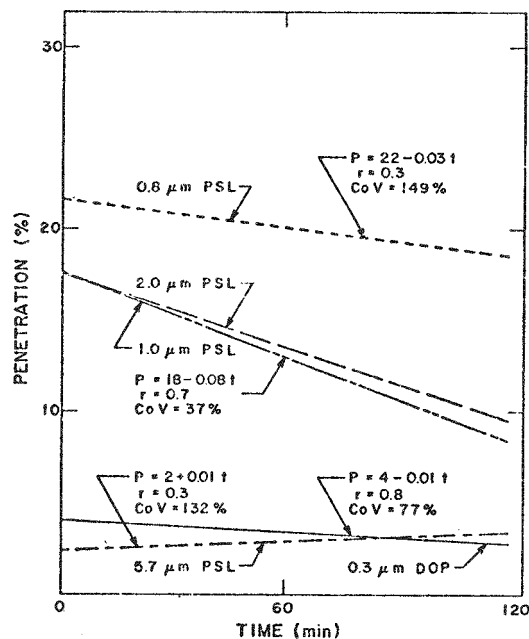


Fig. 15.
Aerosol penetration through G-V-S tubes at 200 cm^3/min .

were the only components tested alone (step 3 of the above experimental procedure). Assuming that the two spacers have equal collection efficiencies, the expression for determining that efficiency is

$$C_o (1 - E_{p1}) (1 - E_{p2}) = C_o (1 - E_{op}). \quad (2)$$

C_o is the initial concentration, E_{p1} and E_{p2} are the individual spacer fractional collection efficiencies, and E_{op} is the experimentally determined collection efficiency with only the spacers in the tube. $E_{p1} = E_{p2} = E_p$, so that Eq. (2) reduces to a quadratic in E ,

$$E_p^2 - 2E_p + 1 = 1 - E_{op}. \quad (3)$$

Using $E_{op} = 0.43$ (from Fig. 16), $E_p = 0.25$.

The charcoal granules are in two sections, the first (upstream) of which is twice the mass of the other. Assuming equal collection efficiencies for each of three equal mass sections of charcoal, the collection efficiency E_c of each section is calculated from the measured penetration through the polyurethane plugs plus the charcoal, $(1 - E_{op+c})$, using

$$C_o (1 - E_c) (1 - E_c) (1 - E_p) (1 - E_c) (1 - E_p) = C_o (1 - E_{op+c}), \quad (4)$$

where $C_o (1 - E_c)$ is the mass fraction penetrating the first half of the large charcoal section, $C_o (1 - E_c) (1 - E_c)$ is the mass fraction penetrating the second half of the large charcoal section, etc. Combining terms

$$(1 - E_c)^3 (1 - E_p)^2 = (1 - E_{op+c}). \quad (5)$$

Substituting $E_p = 0.25$ and $1 - E_{op+c} = 0.25$ (from Fig. 16 for tube with glass wool removed) $E_c = 0.24$. Similarly solving for the glass wool collection efficiency using the foregoing values and the overall observed penetration indicates the glass wool collection efficiency, $E_r = 0.64$. Thus, the portions of the G-V-S tubes that are not analyzed exhibit appreciable collection of $1.0\text{-}\mu\text{m}$ particles.

This point can be further emphasized by the following material balance of the particulates entering and leaving the tube,

$$FC_o t = C_o (1 - E_t) Ft + (C_o E_r + C_1 E_c + C_2 E_c + C_3 E_p + C_4 E_c + C_5 E_p) Ft. \quad (6)$$

$C_o \dots C_5$ are the challenge concentrations to each section, F is flow rate, t is time, E_t is overall tube collection efficiency, and E_r , E_c , and E_p are the collection efficiencies for the glass wool, each charcoal section (the charcoal sections are again considered to be three equal sections for

computational purposes), and each polyurethane spacer. The results of this balance are given in Table IX for an entering aerosol containing $1 \mu\text{g}/\ell$ of $1\text{-}\mu\text{m}$ -diam particles. The calculated efficiencies are used to determine the aerosol mass concentration as it enters each section, and the mass of particles deposited in each component. Thus, from the table, approximately 21% of the collected mass is retained by the charcoal. However, this is only 19% of the total particle mass entering the G-V-S tube.

It is evident that the G-V-S particle collection efficiency could be much improved. The variability of penetration through the tubes suggests the addition of a separate component to collect only particulates. This could be a small cassette containing an efficient fiber glass prefilter to collect particles but not affect the passage of gases. The fiber glass filter would have to be analyzed to determine the presence of contaminant on the particles.

Even without redesign of the G-V-S a better contaminant estimate might be obtained if all the tube components were routinely analyzed. However, to initially (or routinely) determine the presence of particles containing the contaminant, only the glass wool need be analyzed in addition to the charcoal. If analysis of only the charcoal is feasible then simply reordering the G-V-S components to place a polyurethane spacer at the inlet, rather than glass wool, will increase the fraction of particles retained by the charcoal sections to approximately 37% of the total particulates ($1.0\text{-}\mu\text{m}$ particles).

These modifications or procedural changes would not impair the gas or vapor collection and would improve particulate collection to provide a better contaminant analysis.

V. REAL-TIME SAMPLER

Work has begun on the development of a real-time, or short time, aerosol size distribution measuring system. As it is envisioned, the unit will sample a dusty environment, fractionate the dust into known aerodynamic size groups, analyze the mass in each group, and display or print either an aerodynamic size distribution, or simply display the mass data for subsequent calculation of the distribution. Sample fractionation will probably be accomplished by impaction or virtual impaction and the mass analysis by β -counting or piezoelectric crystal frequency change. Since a finite time is required to obtain a sample, the collection and data reduction cycle will most likely require up to 10 min. In addition to size distribution, the instrument will measure total sample mass and provide total mass concentration. The unit will be designed and constructed as a portable, self-contained package weighing less than 23 kg.

If a four-stage cascade impactor (CI) is used to fractionate the aerosol, the principal design and fabrication task will be that of mechanically constructing the stages so that the dust impacted on each stage can be analyzed. Although one sensor could be used at each stage, the expense and complexity make it undesirable. More likely, each stage will be moved mechanically to a single sensing head. To provide a product which is not too expensive to manufacture and market, some compromises must be considered. Four levels of instrument complexity are outlined in Table X. As the complexity increases so does the required development and testing time, as well as the price. The sophistication of the instrument system should be geared to its expected use and an acceptable selling price.

The initial experimental work is aimed toward selecting a suitable sample collection and fractionation technique. Impaction is a straightforward, well-characterized technique for obtaining aerodynamic size distribution between $0.5\text{-}\mu\text{m}$ and $10\text{-}\mu\text{m}$ diam. The impactor is simple in construction, and for this application could be small and lightweight. Its main disadvantage is that particles may bounce or be re-entrained from the impaction surface. This overloading phenomenon occurs less readily if the impaction surface is rough or sticky.

Instead of designing an impaction device, initially we will investigate the feasibility of a virtual impaction (VI) separation device. The VI is not as well characterized as the CI; conse-

quently, considerable experimentation will be necessary to determine its suitability. The VI does not suffer from dust loading as does the CI. On the other hand, mediocre size separation and excessive wall losses have been reported in present generation VIs.^{13,14} In principle, both of these drawbacks can be alleviated by improved design of the inlet and the collector orifice. Combining the mechanics of impaction and virtual impaction may also be feasible.

A virtual impaction device has been constructed. This single-stage device has provision for changing the inlet orifice size, the orifice to collector spacing, and the flow rates, and can be used as a combined impaction virtual-impaction separator (Fig. 17). The VI is presently undergoing flow, pressure drop, and particle separation calibrations.

VI. FUTURE WORK

- Test our fibrous aerosol generation and characterization system with UICC chrysotile "B" asbestos.
- Complete the fiber-sizing portion of the MRE and Spiral Centrifuge intercomparison series.
- Complete work on a topical report describing our cyclone calibration series conducted against seven different fibrous glass aerosols.
- Design, fabricate, and test a prototype separation device to be employed in the real-time aerosol sampler.

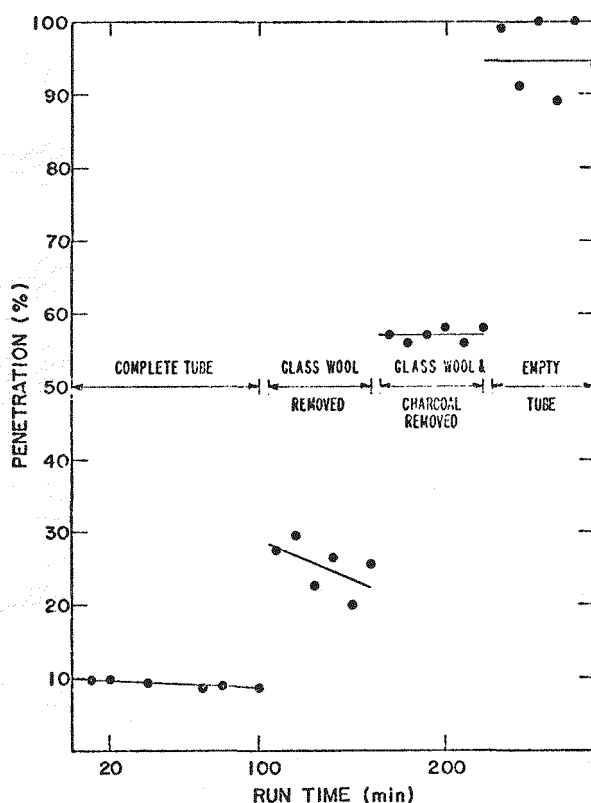


Fig. 16.

Penetration of 1- μ m-PSL aerosol through G-V-S tube components.

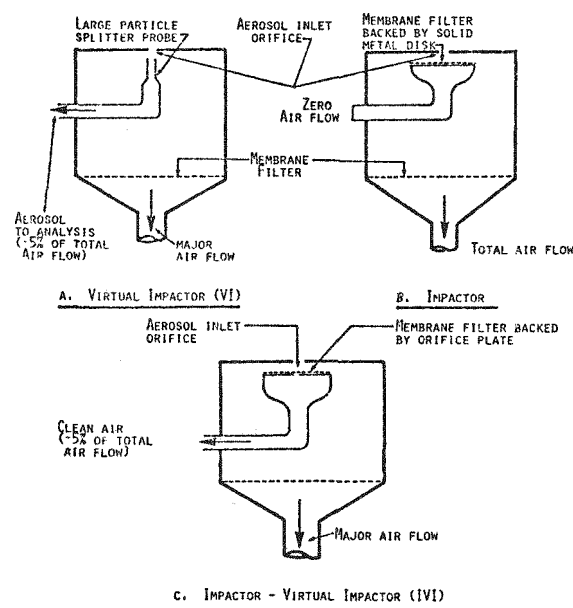


Fig. 17.

Real-time sampler size selector concepts.

TABLE IX
G-V-S TUBE MATERIAL BALANCE

G-V-S Component	Collection Efficiency E(%)	Conc Entering Section ($\mu\text{g}/\ell$)	Mass Deposited ($\mu\text{g}/\ell$)	Percent of Total Deposited
Fiberglass	64	1.0	.64	70.3
1st charcoal	24	0.36	.09	16.7
2nd charcoal		0.27	.06	
1st p.u. plug	25	.21	.05	5.5
3rd charcoal	24	.16	.04	4.4
2nd p.u. plug	25	.12	.03	3.3
Total collected			.91	100.2
Mass penetrating			(.09)	
Total particulates collected in charcoal			.19	21

TABLE X
REAL-TIME AEROSOL SAMPLER OPTIONS

System Description	Example Components
Option 1. Four-stage separator interfaced with commercial single stage detector; commercial data reduction device.	Manually shifted four-stage CI or VI interfaced to GCA RDM or TSI PMM; HP-25 calculator.
Option 2. Four-stage separator with internal single stage detector and display; commercial data reduction device.	Manually shifted four-stage CI or VI with β sources and counter; HP-25 calculator.
Option 3. Four-stage separator with internal single stage detector; internal data reduction and display.	Manually shifted four-stage CI or VI with β source and counter; programmed logic and display.
Option 4. Four-stage separator with internal single stage detector; internal data reduction and print-out.	Automatically shifted four-stage CI or VI with β source and counter; programmed logic and print-out.

REFERENCES

1. Recommendations of the British Medical Research Council Panels Relative to Selective Sampling, March 1952, in **Inhaled Particles and Vapors**, p. 475, Pergamon Press, London (1961).
2. C. I. Fairchild, L. W. Ortiz, H. J. Ettinger, and M. I. Tillery, "Aerosol Research and Development Related to Health Hazard Analysis. July 1, 1974-June 30, 1975," Los Alamos Scientific Laboratory report LA-6277-PR (March 1976).
3. V. Timbrell, A. W. Hyett, and J. W. Skidmore, "A Simple Dispenser for Generating Dust Clouds from Standard Reference Samples of Asbestos," *Ann. Occup. Hyg.*, Vol. 11, 273-81 (1968).
4. L. W. Ortiz, H. J. Ettinger, C. I. Fairchild, and M. I. Tillery, "Aerosol Research and Development Related to Health Hazard Analysis. January 1 through June 30, 1974," Los Alamos Scientific Laboratory report LA-5797-PR (November 1974).
5. Committee on Threshold Limit Values: Threshold Limit Values of Airborne Contaminants for 1968. American Conference of Governmental Industrial Hygienists, Cincinnati, Ohio (1968).
6. H. J. Ettinger, C. I. Fairchild, L. W. Ortiz, and M. I. Tillery, "Aerosol Research and Development Related to Health Hazard Analysis. January 1 through June 30, 1973," Los Alamos Scientific Laboratory report LA-5359-PR (July 1973).
7. V. Timbrell, "The Inhalation of Fibrous Dusts," in **Biological Effects of Asbestos**, *Ann. N. Y. Acad. Sci.*, Vol. 132, Art. 1 255-273 (December 31, 1965).
8. H. J. Ettinger, C. I. Fairchild, O. R. Moss, and L. W. Ortiz, "Aerosol Research and Development Related to Health Hazard Analysis, January 1 through March 31, 1972," Los Alamos Scientific Laboratory report LA-4936-PR (April 1972).
9. R. N. Mitchell, D. A. Bevis, and E. C. Hyatt, "Comparison of Respirator Filter Penetration by Dioctyl Phthalate and Sodium Chloride," *AIHA J.* 32, 357-364, (1971).
10. O. G. Raabe, "The Generation of Aerosols of Fine Particles," in **Fine Particles, Aerosol Generation, Measurement, Sampling, and Analysis**, p. 70, B.Y.H. Liu, Ed., Academic Press, Inc., New York, 1976.
11. R. D. Hiebert, R. G. Stafford, and L. L. Pollat, "Improved Light Scattering Photometer for Air Filtration System Studies," Los Alamos Scientific Laboratory report LA-4627-MS, 7 pages (1971).
12. J. G. Olin, G. J. Sem, and O. L. Christenson, "Piezoelectric-Electrostatic Aerosol Mass Concentration Monitor," *AIHA J.* 32, 209-220 (1971).
13. R. F. Hounam and R. J. Sherwood, "The Cascade Centripeter: A Device for Determining the Concentration and Size Distribution of Aerosols," *AIHA J.* 26, 122-131 (1965).
14. B. W. Loo, J. M. Jaklevic, and F. S. Goulding, "Dichotomous Virtual Impactor for Large Scale Monitoring of Airborne Particulate Matter," Lawrence Berkeley Laboratory report LBL-3854, 40 pages (1975).