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REMOVAL OF RADIOACTIVE AEROSOLS ON HIGH EFFICIENCY FIBROUS FILTER MEDIA

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OAK RIDGE NATIONAL LABORATORY
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

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JUNE 1970

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REMOVAL OF RADIOACTIVE AEROSOLS ON HIGH EFFICIENCY FIBROUS FILTER MEDIA

R.J. Davis, J. Truitt, J. S. Gill, and R. E. Adams

ABSTRACT

One objective was to observe filtration efficiencies of commercially available, high efficiency fibrous filter media to smokes of those materials possible and under environmental conditions expected in water-cooled reactor containments during a severe accident. A second objective was to provide additional understanding of the performance of high efficiency fibrous filters.

The basic experiments were filtration efficiency tests in which aerosol was generated in an electric arc between an activated stainless steel (or zironium) tube filled with an insert of UO_2 and tungsten. The aerosol was pumped through a filter pack which contained three filters in series. The filtration efficiency was taken to be the ratio of the activity on the first filter to the total on all three filters. Filter efficiency was measured for eight different commercial media vs velocity (3.5-10 ft/min), humidity (zero to 100% relative humidity) and temperature (25, 60 and 100°C). Other filtration efficiency tests were done to compare the measurement with the standard DOP test, to determine possible differences in filter efficiency with SS-UO_2 vs ZrO_2-UO_2 particles, and to determine possible effects of water condensing on the filter fibers.

Subsidiary experiments were done to estimate the distributions of fiber diameters (microscopically) for four available media. Other subsidiary experiments were done to estimate the particle size distribution in the arc-generated aerosol with electronic sizing and counting instruments and with a low pressure impactor.

The basic experiment was shown to measure filter efficiencies in the range near 99.97% to $\pm$ 0.01-0.02. The method was shown to give efficiency values in good agreement with those measured with 0.3 micron DOP particles. Penetrations were the same for SS-UO_2 aerosol as for ZrO_2-UO_2 aerosol. The eight different commercial media provided similar efficiencies under all conditions in which comparisons were made. The effects of increasing velocity (from 3.5 to 10 ft/min), of increasing the temperature (from 25 to 100°C), of increasing humidity (from zero to 100% relative humidity) or of steeping the media in saturated, 80°C steam and air prior to use, were to increase the penetration by factors of 1.5 to 2.

The subsidiary experiments to characterize the filter media indicated that [illegible] of the fibers were of diameters near 2 microns or near 0.15 micron. The large fibers apparently hold the mat together; the tiny fibers are largely responsible for the filtering action.

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The subsidiary, particle sizing experiments showed that the arc-generated aerosols contain chain agglomerate particles (i.e., clumps of tiny, usually spherical, nuclei). The diameters of the nuclei (or primary particles) were in the range of a few hundred Angstroms, the length of the agglomerates were in the range 0.1 to 1 micron. The particle size distribution upstream and downstream of a filter were measured with a low pressure impactor. The two size distributions were similar.

Approximate values of filtration efficiency (vs particle size) were calculated using available filtration theory and using the fiber diameter data observed. The approximate effects of velocity, moisture on the particles, moisture on the fibers, temperature and particle density were estimated; the calculated effects were qualitatively similar to the observed effects.

The idea that most of the penetration through an absolute filter is through imperfections or holes, is discussed. The observed similarity in particle size distribution before and after a filter is evidence that holes are responsible for most of the penetration.

I. Objectives

The central issue in nuclear safety is the estimation of the radiological dose that a bystander could receive in the event of a severe accident. Among the information needed for such an estimation is the probable filtration efficiency of a containment filter system under accident conditions. Among the information needed to estimate the efficiency of such a system is the efficiency of the filter media used to aerosols of the kinds expected and under environmental conditions expected.

The major objective of this work was to observe the filtration efficiency of several commercially available, high efficiency, fibrous filter media, to smokes of those materials possible in accidents with water-cooled reactors, under environmental conditions expected in use, in the containment vessels of such reactors. A second objective was to provide additional understanding of the performance of high efficiency fibrous filters so that interpolation and modest extrapolation of observed filter efficiency data can be confidently made.

In order that this work be presented in a useful perspective, it is important first to note some important considerations not included in our objectives. We did not consider leakage, (the importance of sealants, mountings, sudden pressure pulses, etc.) or plugging with either condensed moisture or too much particle phase. These considerations are properly the concern of tests with standard filter units and experience with maintenance of existing systems. We did not consider variability of media, either from spot to spot in a particular sample or from one batch of media to the next. These considerations are more properly the concern of quality control stations. We did not consider the quantitative relation between maintenance schedule, and the probability of leaks of various sizes (i.e., the estimation of the probability that a system will be 90% efficiency, 99% etc. as a function of maintenance).

II. Summary

The basic experiment was a test in which a radioactive aerosol was pumped through a series of three samples of the filter media being tested. The filter efficiency was taken to be the ratio of activity on the first filter to the total activity on all three filters. This basic experiment was done for a variety of commercial filter media, at a variety of

face velocities with ZrO_2-UO_2 and with stainless steel oxide-UO_2 aerosols.[1-5]

Subsidiary experiments were done to characterize the aerosols (i.e., determine particle size) and to characterize the filter media (i.e., to determine fiber diameter).

The experimental procedures for the basic filtration tests, for the fiber diameter measurements and for the particle sizing experiments are each described in the next section. Data and results for each kind of experiment follow the procedures. Finally, a discussion of the results is presented.

III. Experimental Procedures and Equipment

A. Filter Efficiency Measurements

1. The Tank and Piping Arrangement

The general arrangement is indicated in Fig. 1 which is a schematic diagram of the apparatus and Fig. 2 which is a photograph of the same. The general procedure was as follows: the (100-liter) tank was filled with concentrated aerosol by circulating air through the electric arc aerosol generator for about one minute. The arc loop was then valved off and circulation through the filter pack loop was started and continued for about 15 minutes. Circulation was then stopped, the two filter packs were replaced with a fresh pair and circulation was continued for about 30 minutes. This sequence constituted a run and supplied four independent measurements of filter efficiency of a single kind of media.

The flow rates through both filter packs together, and through each individually, were measured with rotameters and adjusted with the throttle valves downstream of the filter packs. The flows were adjusted to achieve the velocity desired through the one-inch diameter (about 5 cm^2 area) circular face of media exposed:

Face Velocity		Volume Flow Through Each Pack
ft/min	cm/sec	(cm^3/sec)
3.5	1.8	9.0
5.	2.5	12.9
7.5	3.8	19.3
10.	5.1	25.8

The pressure drop across each filter pack was monitored with water filled manometers. The pressure drops were always in the range 2-4 cm of water for the conditions reported.

In tests with dry aerosol the relative humidity was less than 5%, as measured at room temperature. In tests with wet aerosol, liquid water was maintained in the bottom of the tank.

The tank and the insulated box containing the aerosol generator, filter packs and piping could be thermostated at elevated temperature. The 25°C runs were done with the whole system at room temperature. The 60 and 100°C runs were done with the tank temperature about 5°C lower than the temperature of the box containing the rest of the system; this was to prevent, in the cases of the wet runs, gross condensation in the filter packs and clogging of the filter media.

The pump was a Leiman Bros. (Newark, N.J.), Rotary Oil-Less Model 202-C pump run at 1725 rpm.

2. The Filter Packs and Analysis

The disassembled filter pack is shown in Fig. 3, and the filter packs are shown in the assembly in Figs. 1 and 2. The filter pack was a stainless steel, cylindrical holder (1-1/2 inch in internal diameter) containing a series of three disks (1-1/2 in. total diameter with a circular area one inch in diameter exposed to flow) of filter media in series. Each filter media disk was supported by a screen; a neoprene O-ring was pressed down onto the filter media by means of special stainless steel rings. The steel rings were followed by a second series: screen, filter media, O-ring and steel rings. This was followed by a third and similar series. Finally the spring was put in, compressed and locked into place. The flow was such that the last filter disc loaded into the holder saw the aerosol first (and will be referred to as the first filter).

The unloading was simply the reverse of the assembly. The relative amounts of aerosol collected on each filter media sample was measured by counting the gamma-ray activity of ^{51}Cr in the activated stainless steel (or ^{94}Zr-^{94}Nb in the activated Zr) used to make the aerosol. The counting was done with a single channel analyzer and a CsI crystal detector. The pieces of the filter pack were counted as follows: The first filter paper (in a light weight metal salve box) was placed on a mount which positioned the sample 10 cm above the crystal and was counted for 10 min. The 10 cm distance was shown experimentally to reduce the count by a factor of 14.86; the observed count was therefore multiplied by this factor to get the equivalent count at the crystal face. The equivalent count on the first filter was typically in the range 100,000 to 300,000. The screen, the retainer rings and O-ring behind the first filter were similarly placed in a salve box, placed directly on the crystal detector and counted 10 min. Similarly, the second filter, the assembly of parts behind the second filter (i.e., screen, steel rings and O-ring), the third filter and the screen behind the third filter were counted. Background counts on all the assembly parts were measured before the experiment and subtracted from the counts with aerosol deposit to get net values.

The filter efficiency was taken to be the ratio of the counts found on the first filter to the total counts on all filters and assembly parts downstream of the first filter.

The typical amount of material collected on the first filter was too small to weigh (i.e., less than a few tenths of a milligram). No significant pressure drop increase during the experiments was ever observed.

There were two types of experimental difficulties encountered with this system. One resulted in the occasional occurrence (about one in 10-15) of very low filter efficiency values. It is presumed that in these cases some extra material was transferred to the second or third filter during the unloading procedure. The practice of always making four measurements of each efficiency value allowed these occasional, extraordinary values to be confidently discarded.

The second difficulty arose with leaks in those experiments in which condensation could occur in the filter packs. These were the experiments run with whole circulation system at room temperature and with liquid water standing in the bottom of the tank. The leaks were worst in the special sequence of experiment in which (in addition to humidity from water in the tank) the media samples were pretreated by steeping them in saturated, 80°C steam-air for 24 hrs prior to test. This problem was adequately solved in the cases of tests with dry media and wet, 25°C aerosol by repeating the experiments and discarding, on statistical grounds, the low efficiency values. In the cases of the special sequence of tests with media prepared by steeping in 80°C water-saturated air, the leaking was avoided by the tedious technique of gluing the media into the holder with Duco cement.

3. Aerosol Generator

The aerosol generator was an electric arc between tungsten and a 1/4-in. O.D., 1/16 in. I.D. stainless steel (or Zircalloy-2) tube filled with a cylinder of compacted UO_2. A photograph of such an arc, after brief use, is shown in Fig. 4. It is clear that the UO_2 as well as the stainless steel was consumed. The electrodes were mounted in a one-inch glass pipe tee as shown in Fig. 2. The arc was powered with a commercial arc welding unit.

B. Particle Size Measurements

1. By Electronic Counting Instruments

To generally characterize the arc generated aerosol the following experiment was performed by McFarland et al.[6] at the University of MinnesotaParticle Laboratory. An aerosol was generated by an electric arc between two stainless steel rods and then transported into a plastic bag to allow agglomeration to take place. Aerosol was drawn from the bag into an analytical system composed of three measuring instruments: a Royco Model 202 optical counter[7], a University of Minnesota electrostatic particle counter[8] and a General Electric condensation nuclei counter.[9] The optical counter indicated the number of particles in each of a succession of size ranges from 0.3 micron to several microns. The Minnesota counter indicated the number of particles in size ranges in the overall range from about 0.07 to 0.3 micron. The nuclei counter gave an indication of the total concentration of all particles from a few thousands of a micron to larger; this total concentration minus the concentration of particles 0.15 micron and larger provided an indication of the number of particles from about 0.01 to 0.07 micron.

The size indications from these instruments should be taken as approximate.

2. By the ORNL Low Pressure Impactor

This instrument has been described in detail elsewhere.[10] It measures particle diameter (assuming the particle density is 5 g/cm^3) in the range 0.01 to 2 microns. Two experiments were done with the impactor. In both experiments the 100-liter tank was filled with SS-UO_2 arc generated aerosol (as described above for filtration efficiency measurements). The filter packs in the system shown in Fig. 1 were removed to provide two open pipes into the system. In one experiment the impactor was piped onto one opening and a filter was installed on the other. The 100-liter tank was filled with aerosol as in the filtration tests, then the impactor was operated for 15 min or so at the required flow rate of 8 liters (STP)/min. The relative amounts of material on the impactor stages and the after filter in the impactor were measured in terms of gamma-activity of ^{51}Cr (as in the filtration efficiency tests).

In the other experiment, a large filter was placed between the aerosol system and the impactor. In this experiment the 15 min circulation through the impactor was followed by a second aerosol generation period to refill the 100-liter tank. This process was repeated 8 more times in order to get enough aerosol through the filter and into the impactor to count. The filter media used was Flanders 700. The face area of the filter was about 123 cm^2 (12.5 cm diameter). At 9 liters/min flow the face velocity was about 1.1 cm/sec or 2.3 ft/min.

C. Fiber Diameter Measurements

Two types of fiber diameter measurements were made for samples of four different filter media (Flanders 800, AAF-1, Cambridge 115E and MSA Ultra Hepa). In one examination the filter media were dispersed in water by use of an ultrasonic generator and drops of the suspension were put onto glass slides, viewed under an optical microscope and photographed. The numbers of fibers in each of several fiber diameter increments were noted. In the second examination, samples of the filter media were mounted in butyl-methyl methacrylate and sections of the mount, including fibers in cross section, were cut with an ultramicrotome and viewed under an electronmicroscope. Again fiber counts versus fiber diameter were made.

IV. Data and Results

A. Filtration Efficiencies

1. Reproducibility of the Measurement

In order to demonstrate the reproducibility of the measurements, a series of 16 values of filter efficiency was determined for one media (Flanders 700) at standard conditions (i.e., 5 ft/min with dry SS-UO_2 aerosol at 25°C). These values are listed in Table 1.

Among the 16 values (of percent of substance filtered) there was one extraordinarily low value. As mentioned earlier, the occurrence of an occasional low value was typical. It was presumed to have resulted from contamination of one of the two downstream filter samples or of the assembly parts during unloading of the filter pack. The low value was therefore discarded.

The other 15 values agree well with each other, with an average value of 99;97% filtered, an average deviation from the average value of 0.01 (% filtered) and a total range of values of about ± 0.02 (% filtered).

The precision of the values of % filtered or of % penetration is indicated to be about ± 0.02.

2. Comparison with the DOP Test

It is common practice to test the filtration efficiency of filter media with "DOP hot smoke".[11] DOP is an oil (dioctyl phthalate). The term, "hot smoke" indicates a method of particle generation in which dioctyl phthalate vapor is condensed onto nuclei under controlled conditions to give a nearly monodisperse aerosol of 0.3 micron diameter droplets.

A comparison was made of filtration efficiency to the arc-generated, solid, agglomerate particles of SS-UO_2 with filtration efficiency to DOP particles. Samples were obtained of two media which had been tested for DOP penetration (as well as other parameters)[12] at an AEC Quality Assurance Station. The results of those tests are given in Table 2 along with filter penetrations by wet and dry SS-UO_2 aerosol which we measured.

The agreements between the penetration values are well within the precision of our measurement; penetration values for the two aerosols are the same.

3. The Effects of Velocity, Humidity, and Temperature

Figures 5 is a plot of percent penetration vs velocity (3.5, 5, 7.5 and 10 ft/min) for several commercial media to dry (< 5% relative humidity) stainless steel oxide-UO_2 aerosol at 25°C. Figure 6 is a similar set of results for wet (approximately 100% relative humidity) aerosol.

Figures 7 and 8 are plots of penetration vs velocity (5 and 10 ft/min) for dry and wet aerosol, respectively, at 60°C. Figures 9 and 10 contain similar data for 100°C.

Since there were no differences in penetration from media to media (and small differences are insignificant in as much as samples from different batches, different parts of the roll etc. were not tested), the penetration values for all the media were averaged to show more precisely the effects of temperature, velocity and humidity. These averages are given in Fig. 11. The penetrations increase by small amounts with increasing velocity, with increasing temperature and with increasing humidity.

4. Comparison of Stainless Steel Oxide-UO_2 and ZrO_2-UO_2 Aerosols

A long series of runs was made with ZrO_2-UO_2 aerosol at 25°C both dry (< 5% relative humidity) and wet (approximately 100% relative humidity). The results are compared with corresponding results with stainless steel oxide-UO_2 in Table 3.

The agreements between penetration values for the two aerosol materials are in all cases well within the precision of the measurement.

5. The Effect of Water on the Fibers

Gross amounts of condensation will plug a filter, somewhat less will cause large pressure drops. These effects are known and understood.

It is conceivable (as described in detail later) that more modest amounts of water could drastically decrease filter efficiency without increasing the pressure drop. We wished to test this possibility. Samples of filter media were stored in a dessicator (which had water in it) in an oven at 80°C for 24 hrs then subjected to wet (SS-UO_2) aerosol at 25°C. At 80°C, water saturated air is about half water; so these media samples were steeped in concentrated and saturated water vapor for 24 hrs to allow adsorption of water on the fibers.

In other tests the media samples were steeped in the water vapor then dried at 100°C before test to see if any loss in filter efficiency would be restored by drying.

The results are given on Table 4. The results show a small increase in penetration due to steeping in water vapor. It is not clear whether the drying caused the filter efficiency to recover or not.

B. Characterization of the Aerosols

1. Electron Photomicrographs

To characterize the shapes of particles of interest, several electronphotomicrographs are presented. Figure 12 is a picture of a dry, stainless steel oxide aerosol generated in an electric spark between stainless steel and tungsten. This sample was precipitated directly onto an electronmicroscope grid mounted in a thermal precipitator. It shows the chain agglomerates, generally of lengths in the range 0.1 to 1 micron.

Figure 13 is a closeup of an agglomerate from an aerosol formed in an arc between a stainless steel tube with a UO_2 insert and tungsten in dry air. The sample was collected on a membrane filter onto which a carbon film had been evaporated. The membrane was later dissolved away, the carbon film picked up on a grid and observed. The primary particle diameters (i.e., the diameters of the individual spheres) vary from about 0.01 to 0.07 micron.

Figure 14 is a picture of a particle from a similar aerosol except the air was wet (> 90% relative humidity). The sample preparation was via the membrane filter technique.

Figure 15 is a picture of a particle from an aerosol formed in an arc between a stainless steel tube with a ZrO_2 insert and tungsten in dry air. The primary particles are less perfect spheres in this case.

Figure 16 is a picture of some primary particles from an aerosol formed in an electric arc between uranium metal and tungsten in wet

(> 90% relative humidity) air. There are several flat sides here, which indicates crystallization.

2. Uranium Content of Stainless Steel Oxide-UO_2 Aerosols

It was shown by several measurements of the ^{51}Cr activity (to indicate stainless steel content) and ^{235}U activity (to indicate UO_2 content) in aerosol samples deposited on filters that the uranium-to-steel-weight ratio was about 0.01-0.03. The uranium-to-steel-weight ratio in the electrode was nearly unity.

3. Particle Size Distribution from Electronic Counting Instruments

The instruments provided values of ΔN, the number of particles in each of a succession of particle radius ranges of width ΔR and average value R. This data is particle number distribution data; it was transposed to particle volume fraction distribution (i.e., the relative fraction of the volume of particle phase in each particle radius range was particle radius) as follows.

The volume of particles per cm^3 in each radius range was calculated

$\Delta\phi = 4/3\pi R^3(\Delta N)$.

The total volume of particles in one cm^3 was calculated:

$\phi = \Sigma\Delta V$.

ϕ is called the volume fraction. Finally values of $(1/\phi)(\Delta\phi/\Delta R)$ were calculated for each radius range and plotted against the values of R. This plot is given in Fig. 17.

Figure 17 is a normalized distribution function; the integral of $(1/\phi)(\Delta\phi/dR)$ between two values of R gives the fraction of the volume (or mass) of particle phase which is in that particle radius range.

The formula:

$$\frac{1}{\phi}\frac{d\phi}{dR} = 1/[R \ln (RMAX/RMIN)]$$

and the concepts of RMAX and RMIN have been described elsewhere.[17] Since the significance of particle size values indicated by the counting instruments has not been precisely demonstrated, for agglomerates of stainless steel oxide, these results should be taken as approximate.

4. Size Distribution Upstream and Downstream of a Filter from Low-Pressure Impactor Measurements

The ORNL low-pressure impactor provides, for spherical particles, values of the mass of particle phase for a sequence of values of $R^2\rho$. There is some uncertainty about the significance of radius (R) and density (ρ) of an agglomerate. There is evidence, both theoretical[13] and experimental[14], that agglomerates would behave in an impactor approximately like spherical particles of the same volume (i.e., like spheres made by melting the agglomerate into a dense ball). With this concept in mind we presumed the particle density, ρ, to be 5 g/cm^3 which is approximately the density of oxides of stainless steel. With this assumed value of ρ, the median radius values for each impactor stage are 1.8, 0.69, 0.26, 0.10, 0.036 and 0.012 micron. The size distribution of the stainless steel oxide-UO_2 aerosol was such that insignificant (^{51}Cr) counts were found on the first (R = 1.8 micron) and last (R = 0.012 micron) impactor stages. For each experiment, the data were therefore, values of the total number of counts (on all the stages and after filters), the number of counts due to particles in the radius range 0.38 to 1.00 micron with a middle value of 0.69 micron, the number of counts due to particles in the radius range 0.14 to 0.38 micron with a middle value of 0.26 micron, the number of counts due to particles in the radius range 0.055 to 0.14 micron with a middle value of 0.10 micron and the number of counts due to particles in the radius range 0.018 to 0.055 micron with a middle value of 0.036 micron.

As is the above case, it was desired to express the results as a normalized volume fraction distribution function, $\frac{1}{\phi}\frac{\Delta\phi}{\Delta R}$, where ϕ is volume fraction (volume of particle per unit volume of aroosol) and R is particle radius. $\Delta\phi/\phi$ was calculated as the ratio of the counts on one stage to the total counts and $1/\phi(\Delta\phi/\Delta R)$ was then computed by dividing by the radius range (i.e., by 1.00-0.38 or 0.62 for the first stage).

The results of Fig. 18 labeled "unfiltered size distribution" are, in fact, this observed size distribution. Results from four separate experiments are plotted as well as the average of the values (the dashed line) from the four experiments.

The data for the experiments in which particles which penetrated a Flanders 800 medium at a velocity of 2.3 ft/min were similar and were treated similarly with one exception. The total counts (i.e., the value which is proportional to ϕ) was taken to be the total counts on the Flanders 800 media as well as the total counts found in the impactor. The results are plotted on Fig. 18 and labeled "filtered size distribution". The results of these experiments are plotted. The averages of the values from the three experiments are also plotted as a dashed line.

Figure 18 also contains a dotted line labeled "calculated assuming 99.97% filtration efficiency for all sizes". These values were calculated by multiplying the averaged values for the "unfiltered size distribution" by the nominal fraction penetration of a high efficiency (99.97%) filter; namely 0.0003. The general agreement between this dotted line and the data suggest that there is no significant variation of filter efficiency with particle size in the size range of the data.

C. Fiber Diameters

The fiber diameter measurements gave values of number of fibers in each of a succession of fiber diameter ranges. There were two sets of such data; one from the optical microscopy, one from the electron-microscopy. The data were transposed to a number distribution function, i.e., the number of fibers per unit of fiber diameter ($\Delta n/\Delta Dp$) vs fiber diameter (Dp). The numbers of fibers, ΔN, in each diameter range of width ΔDp and middle value Dp were the primary data. $\Delta N/\Delta Dp$ values were calculated and plotted, on Fig. 19, for the optical microscope measurements and on Fig. 20 for the electron microscope measurements.

It can be readily seen that the optical microscope results (Fig. 19) show the existence of a predominance of 2 micron diameter fibers and the electronmicroscope results (Fig. 20) show a predominance (i.e., another peak in the number distribution) of 0.15 micron fibers. There may be a subsidiary peak with two of the media at about 0.35 micron.

V. Discussion

A. Filtration Efficiency vs Environmental Conditions

The first objective was to observe filtration efficiencies of several available media to radioactive aerosols of those materials possible in accidents with water-cooled reactors, under environmental conditions expected in use, in the containment vessels of such accidents. This objective was discharged directly with the data (mostly summarized on Fig. 11). Filtration efficiencies were measured; the technique was shown to be able to measure efficiencies in the range near 99.9% to within about 0.02%. The technique was shown to give filtration efficiency values under room conditions which agreed with those measured with 0.3 micron DOP (dioctyl phthalate) droplets. The test aerosols used were either stainless steel oxide-UO_2 mixtures (1-3 wt % UO_2) or ZrO_2-UO_2 mixtures; they were produced in an electric arc. It is anticipated that the smoke in a reactor accident would be nucleated from a homogeneous, concentrated vapor in a steep temperature gradient similar to the nucleation zone near the arc. It is anticipated that a high concentration of very tiny nuclei would initially form in either case and then rapidly agglomerate to give a distribution of agglomerate sizes similar to that shown in Fig. 12. More quantitative descriptions of the ranges of possible aerosols in accidents is the subject of work in progress. The environmental parameters considered were: velocity through the filter (3.5-10 ft/min), temperature (25-100°C) and humidity. Humidity was presumed to be potentially capable of affecting the aerosol or the filter fibers: the effect on filter efficiency due (mainly) to the effect of water on the particles was indicated by tests with humid aerosol with media samples that were (initially) dry. Potential additional effects due to interaction of water with the fibers was indicated by tests with media samples that had been steeped in about a 50-50% mixture of air and saturated water.

A summary of the ranges of parameters and their effects are given in Table 5. Generally the effects were always a small (a factor of 1.5 to 2) increase in filter penetration on changing conditions from the nominal ones: 25°C, dry, and 5 ft/min. There was no difference due to changing aerosol material from stainless steel oxide-UO_2 to ZrO_2-UO_2.

There were no important special effects noted due to coupling of parameters (i.e., 100°C and high velocity or other combinations).

The limitations of the results should also be mentioned to preserve a useful perspective. The results reported are pertinent to the properties of small samples of the filter media. They do not predict anything about leaks either through holes in sealants, through the mounting frames through inferior spots in the media, through possible large holes (due to gross damage of the filter), through open doors which allow parallel flow around the filter, or other possible leakage. A systematic assessment of leakage may be more important than filter efficiencies, i.e., it may be more probable for a particle to escape the containment by going around the filter rather than through it.

B. Application of Filtration Theory to High Efficiency Filters

1. Two Possible Penetration Mechanisms

Langmuir[15] considered the basic mechanisms by which fibrous filters collect particles. Others, for instance, Torgeson[16], have developed the concepts into rather sophisticated schemes to calculate the collection efficiency of fibrous mats. The basic concepts consider the flow lines around a cylindrical obstruction (i.e., a fiber) and the probabilities of a particle hitting the obstruction: (1) simply because the flow line which the particle is on passes within the distance of a particle radius from the fiber (i.e., interception), (2) because the momentum of the particle causes it to cross flow lines and make a bee line to the fiber (i.e., impaction), and (3) because the zig-zag (Brownian motion) path at the particle happens to carry it to the fiber (i.e., diffusion).

The application of these concepts to a dense mat of fibers is inaccurate because the close proximity of fibers to each other results in the flow lines around each fiber being dependent on neighboring fibers. However, the theory could still be useful as a semi-quantitative guide, i.e., to suggest perhaps that velocity is an important parameters, that temperature is not and etc. It was with this point of view that fiber diameters were measured, approximate filter efficiencies were calculated and theoretical prediction of effects of various parameters was made. These calculations are described below.

Recently an alternate, much less sophisticated concept arose. These filter media are very efficient, only three particles out of 10,000 get through under normal operation. It is reasonable that these three out of 10,000 penetrate the filter because they happen to hit a spot in the filter where there are fewer fibers. In other words, it is suggested that the collection efficiency over most of the filter is (via the Langmuir mechanisms and) much greater than 99.97%. An occasional weak spot allows a few particles through to give the observed efficiencies. The spots would have to be rather small and uniformly distributed.

Validity of this simple notion, that penetration occurs mostly through holes, would have an important implication. It would predict filter efficiency to be independent of particle size. It would suggest that (insofar as media properties are concerned) filters in series would each successively remove (at normal operating conditions) 99.97% of the particles.

2. Estimation of Filtration Efficiencies Via the Langmuir Concepts

(a) Description of the Calculation

The estimation of filtration efficiencies was done with the formulations of Torgeson.[17] The calculation provides values of filter efficiency (of a mat of single sized fibers) as a function of aerosol particle size. Of the several parameters needed, the difficult one to obtain (at least for a high efficiency filter which uses very tiny fibers) is the fiber diameter, or distribution of fiber diameters. The fiber diameters were observed as previously described and shown in Figs. 19 and 20. To a good approximation, the data show two fiber diameters: 2 microns and 0.15 micron. Apparently the 2 micron fibers hold the mat together and the 0.15 micron fibers do most of the filtering.

It was not possible from the fiber diameter data to determine the relative amounts of the two sizes of fibers; it was assumed that the large (2 micron) fibers made up 95% of the weight of the mat and the small (0.15 micron) fibers were the other 5%. Indeed, it was presumed that it would be adequate to consider that the mat acted like two mats in series; one of two micron fibers only and one of 0.15 micron fibers only.

The other parameters are the density of the mat which is commonly about 0.008 g/cm^2 for media tested (i.e., 0.008 g/cm^2 of 2 micron fibers and 0.0004 g/cm^2 of 0.15 micron fibers), the temperature (taken to be 25°C), fiber density (2.4 g/cm^3 for glass), and the velocity (5 ft/min)

The efficiency of each hypothetical filter (i.e., the 2 micron fiber mat and the 0.15 micron fiber mat) were calculated. The combined efficiency of the two hypothetical filters was readily calculated on the assumption that each filter acts independently. The first filter collects a fraction ε_1 and passes a fraction $1 - \varepsilon_1$ of the material. The second filter collects a fraction ε_2 of that which passes through the first filter; that is, $\varepsilon_2(1 - \varepsilon_1)$. The two filters together collect the fraction

$$\varepsilon_{tot} = \varepsilon_1 + \varepsilon_2(1 - \varepsilon_1).$$

These calculated filter efficiencies, ε_1, ε_2, and ε_{tot}, are given as a function of particle size in Fig. 21. The minimum in the combined filter efficiency (ε_{tot}) is 98.5% at 0.2-micron particle diameter. The calculations show that the very small (0.15-micron)fibers are responsible for a major part of the filtering action. (It should be reiterated, at this point, that this calculational technique is not suggested as a means of calculating efficiencies for filters in use; it is being used only as a guide to understanding mechanisms of filtration).

(b) Calculated Effect of Velocity

The above calculation was repeated for a sequence of velocities and the results are plotted on Fig. 22. In a rough way one can see that for an aerosol with particles in the diameter range 0.1 to 1 micron, increasing the velocity for 5 to 10 ft/min would increase the penetration by a factor of about 2-3. (That is, the area above the 5 ft/min curve is 1/3 to 1/2 of the area above the 10 ft/min curve.) This is in reasonable agreement with the observed (x 1.5) effect.

(c) Possible Effect Due to Moisture-Agglomerate Interaction

It has been shown elsewhere[18] that there is a tendency for agglomerate particles to become more compacted in high humidity. The effect is indicated pictorially on Fig. 23. This compaction may decrease the apparent size of the agglomerate. For an aerosol with agglomerate sizes in the range 0.1 to 1.0 micron, this would put more particles into the size range less efficiently filtered (as can be seen in Fig. 22). The magnitude of such an effect would probably be modest which more-or-less agrees with the observed effect of humidity.

(d) Calculated Effect of Temperature

Another series of calculations of filtration efficiency were done at different temperatures: 25, 60 and 100°C. The results (filter efficiency vs particle diameter) are given in Fig. 24. The effect (calculated) of temperature is small and is a slight increase in efficiency with increasing temperature. The observed effect was a small decrease in efficiency with temperature.

(e) Calculated Effect of Particle Density

The calculated effect of particle density is indicated on Fig. 25. 10 g/cm^3 particles (in the size range of the saddle in the curve) are indicated to be filtered 3-4 times more efficiently than 1 g/cm^3 particles.

(f) Possible Effect of Moisture on Fibers

It was postulated that moisture might, under some conditions, condense on the fibers and the surface tension between touching, wet fibers might pull them together into bundles as illustrated in Fig. 26. This would have the effects of reducing the number and increasing the diameters of the small fibers. To indicate the conceivable severity of such an effect it was presumed, as an example, that fiber growth occurred such that all the small (0.15 micron) fibers had doubled in diameter (to 0.3 micron). The comparison of calculated filter efficiencies (of a mat with 5 wt % 0.15 micron fibers with a mat with 5% wt 0.3 micron fibers) is shown in Fig. 27.

The predicted effect would be quite large. The observed results did not indicate any such large decrease in efficiency. Apparently the fiber growth (as in Fig. 26) does not occur to any significant extent.

3. Evidence that Penetration is Mostly Through Holes

If the penetration through a filter occurs via the Langmuir concepts then those particles which penetrate would tend to be in the size range of the filtration efficiency saddle (i.e., a few tenths of a micron in diameter). No such tendency was indicated in the size distribution measurements with the low-pressure impactor (Fig. 18).

It is (probably) not possible to make an exact prediction of filtration efficiency (due to Langmuir collection concepts) for high efficiency filters because of the disturbance of the air flow around fibers due to the close proximity of neighboring fibers. It is not possible therefore to be certain that the lack of an observable change in the particle size distribution is proof that penetration is mostly by some other mechanism (i.e., other than the Langmuir concepts). These data do suggest another mechanism; penetration through weak spots or holes would fit the observation. In any case, the data indicate directly that there is no particular particle size range that is filtered at a very low efficiency (i.e., there is no slippery particle size that is unfiltered).

Table 1. Reproducibility of Filter Efficiencies
(Flanders 700 paper, 5 ft/min, dry atmosphere)

% Filtered	% Penetration	Dev. from Avg.
99.990	0.010	-0.023
99.987	0.013	-0.020
99.983	0.017	-0.016
99.974	0.026	-0.007
99.974	0.026	-0.007
99.971	0.029	-0.004
99.968	0.032	-0.001
99.966	0.034	+0.001
99.965	0.035	+0.002
99.963	0.037	+0.004
99.960	0.040	+0.007
99.958	0.042	+0.009
99.954	0.046	+0.013
99.950	0.050	+0.017
99.948	0.052	+0.019
99.680*	0.320*	*
Avg. 99.97	0.03	± 0.01
Range		± 0.02

*Discarded.

Table 2. Comparison of Filter Penetration by Stainless Steel-UO_2 and by DOP Aerosols

Media Identification	Velocity (cm/sec)	Penetration (%) 5.2	14.2
	DOP AEROSOL, 25°C, DRY		
F700		0.021	0.054
H93E		0.021	0.055
	SS-UO_2 AEROSOL, 25°C, DRY		
F700		0.030	0.042
H93E		0.033	0.047
	SS-UO_2 AEROSOL, 25°C, WET		
F700		0.025	0.052
H93E		0.026	0.051

Values of Other Parameters	F700	H93E
Resistance (mm H_2O at 5.2 cm/sec)	31	34
Resistance (mm H_2O at 14.2 cm/sec)	83	90
Tensile strength (in machined direction, #/in.)	5.3	4.8
Tensile strength (in cross machine direction, #/in.)	3.4	3.6
Caliper (inches)	0.018	0.018
Basis weight (#/3000 ft^2)	48.8	57.2
Wt. % combustible	0.7	4.5
Wt. % moisture	0.5	0.5

Table 3. Comparison of Penetration by SS-UO_2 and ZrO_2-UO_2 Particles

Media	Penetrations (%)			
	Dry		Wet	
	SS-UO_2	ZrO_2-UO_2	SS-UO_2	ZrO_2-UO_2
Cambridge 115*	0.102	0.086		0.086
Cambridge 115 WP	0.050	0.047	0.046	0.051
MSA Ultra Hepa	0.026	0.034	0.021	0.043
AAF Type 57		0.032	0.026	0.028
AAF 1	0.031	0.030	0.033	0.031
Flanders 600*	0.078	0.045		0.057
Flanders 800	0.025	0.034	0.025	0.028
Flanders 700	0.036	0.029	0.029	0.028

Velocity: 5 ft/min

Temperature: 25°C

*Non-waterproof media.

Table 4. Effects of Water Condensed on Fibers on the Penetration of Filter Media by Aerosol

Media	Penetration (%)		
	Dry Filter Mat	Over 80°C Water 24 hrs	Mat Dried at 100°C
Cambridge 115		0.088	
Cambridge 115 WP	0.057	0.085	
Flanders 800	0.053	0.087	0.079
Flanders 700	0.045		0.051

Aerosol velocity: 10 [illegible]/min

Temperature: 25°C

Table 5. Summary of Effects of Environment on Filter Penetration

Parameter	Range	Effect on Penetration
Velocity	3.5-10 ft/min	x 2
Temperature	25-100°C	x 1.5
Humidity on particles	0-100% R.H.	x 1.5
Humidity on fibers	Room conditions vs saturated at 80°C	x 1.5
ZrO_2-UO_2 vs SS-UO_2	25°C, 5 ft/min wet and dry	None

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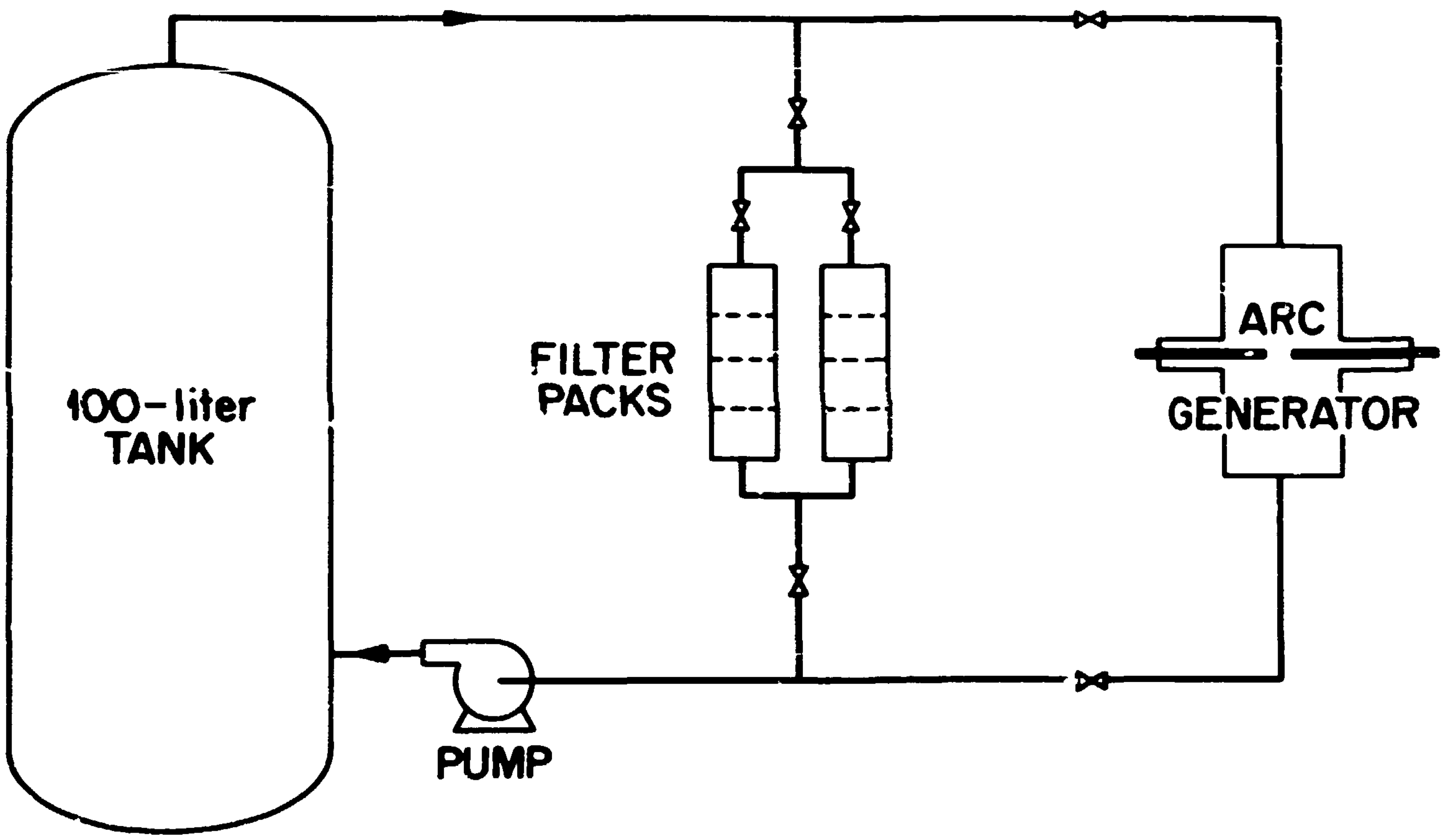

Fig. 1. Schematic Diagram of Apparatus Used to Test Efficiencies of Samples of Filter Media.

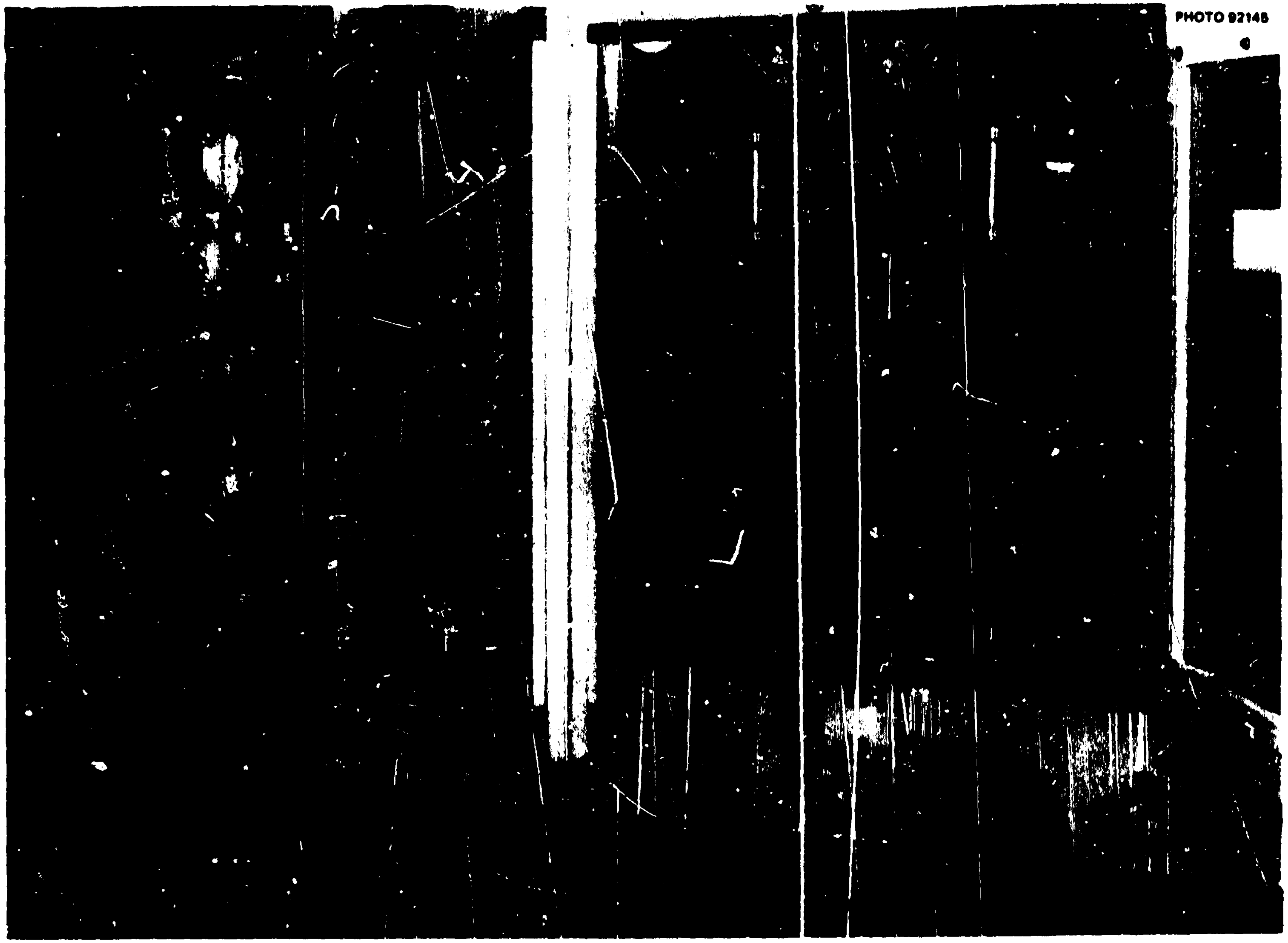

Fig. 2. Apparatus Used to Test Efficiencies of Samples of Filter Media.

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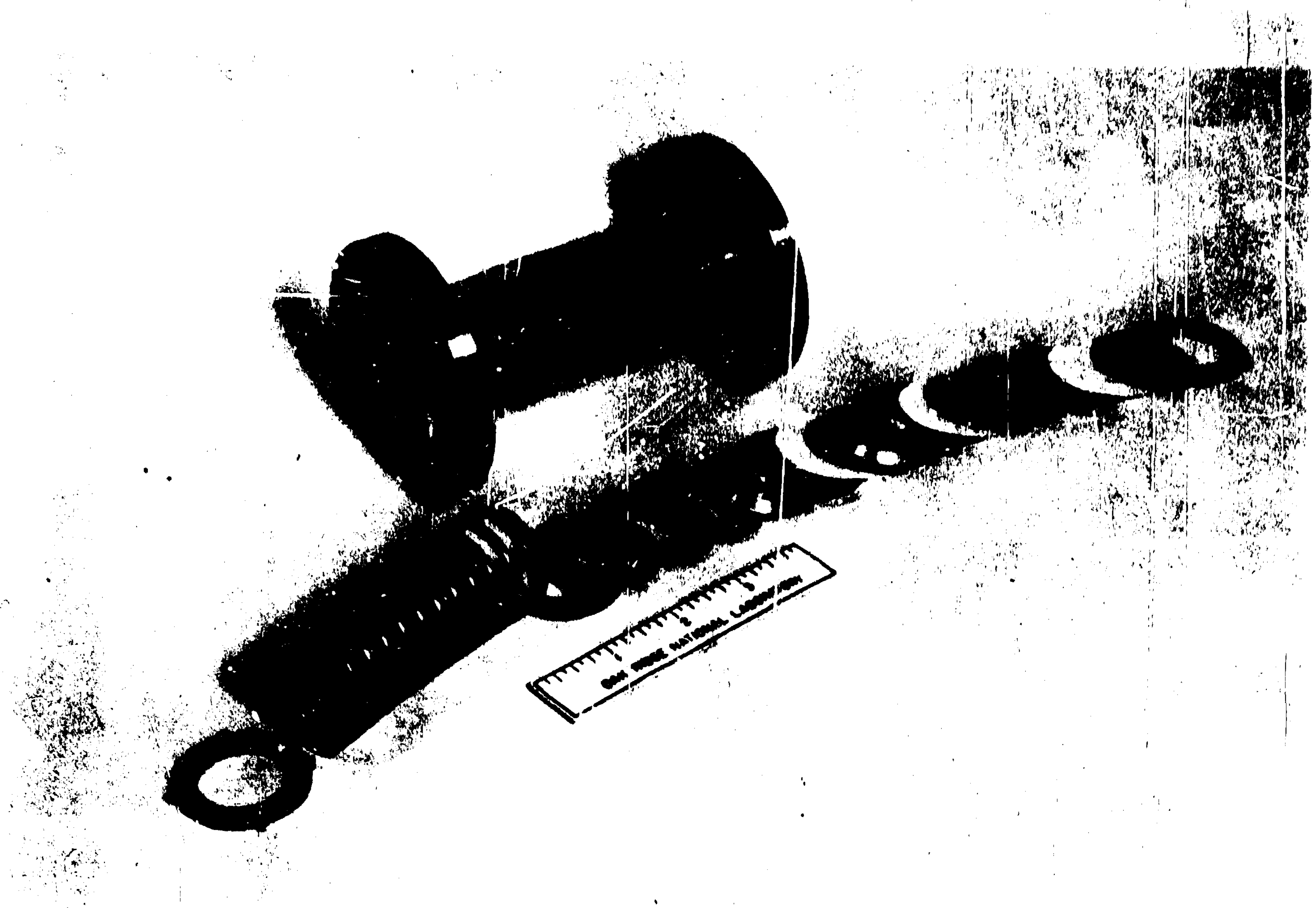

Fig. 3. Disassembled Filter Pack.

Fig. 4. Stainless Steel-UO_2 to Tungsten Electric Arc Aerosol Generator.

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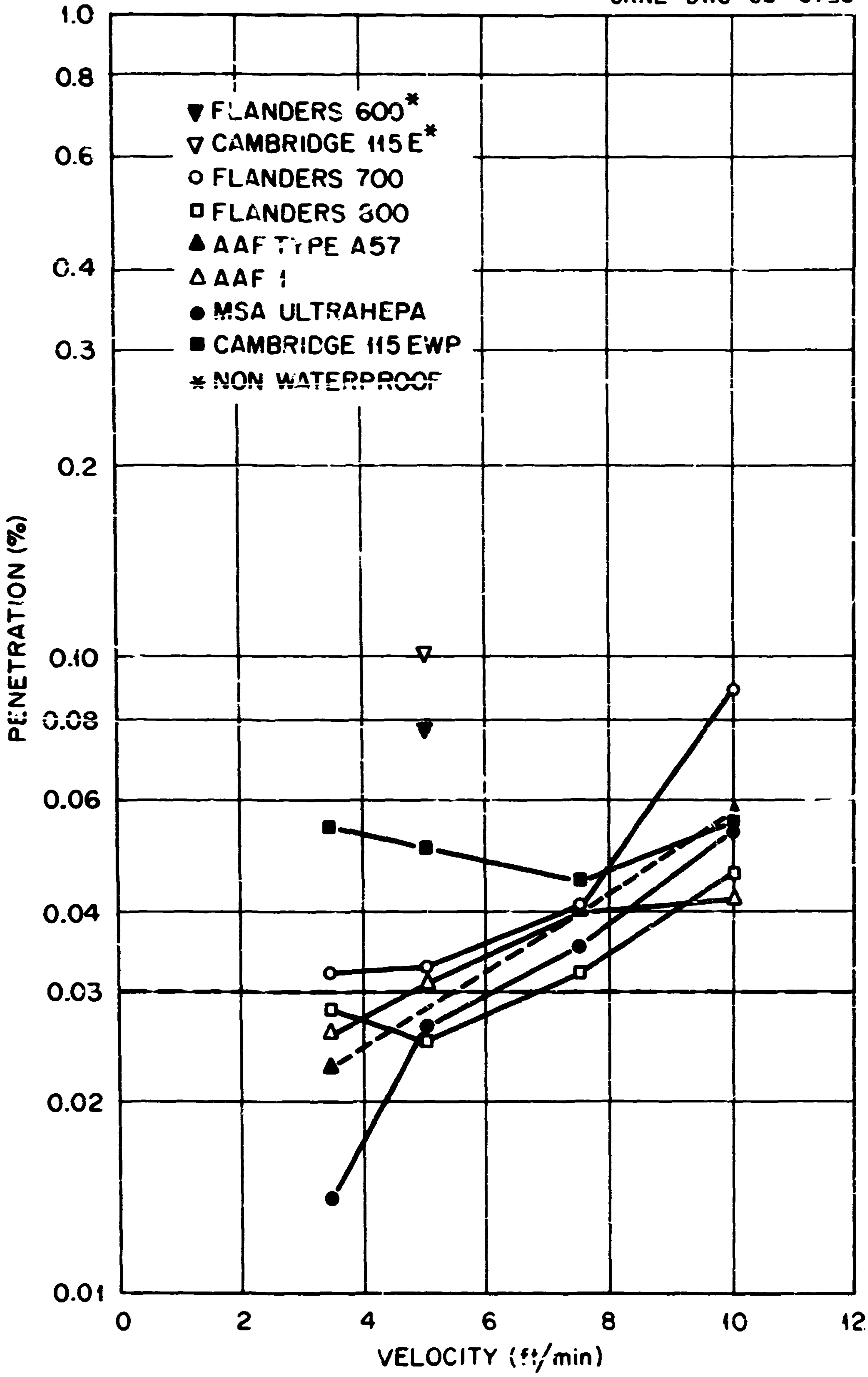

Fig. 5. Filter Efficiencies - Dry Stainless Steel-UO_2 Aerosol at 25°C.

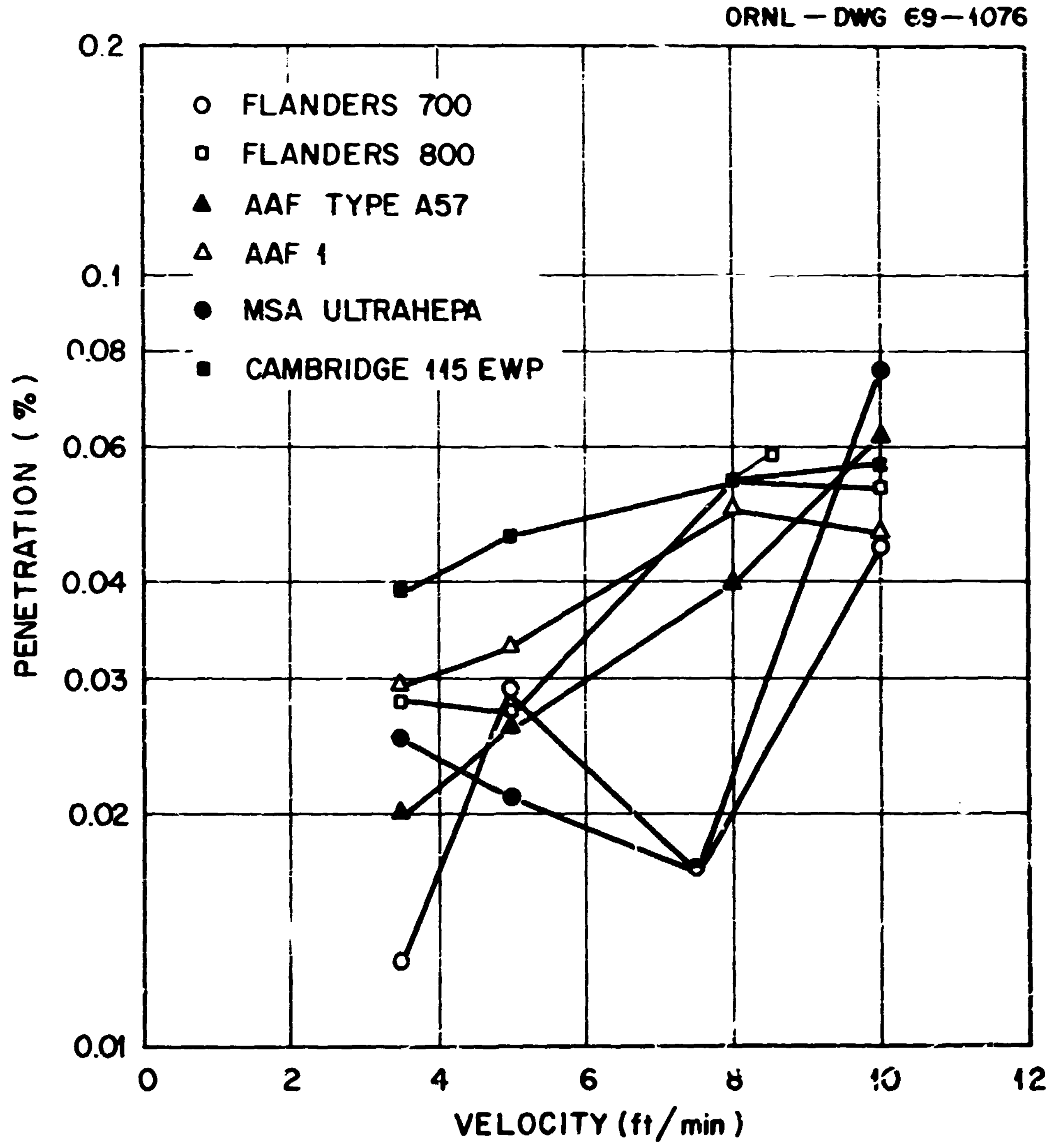

Fig. 6. Filter Efficiencies - Wet Stainless Steel-UO_2 Aerosol at 25°C.

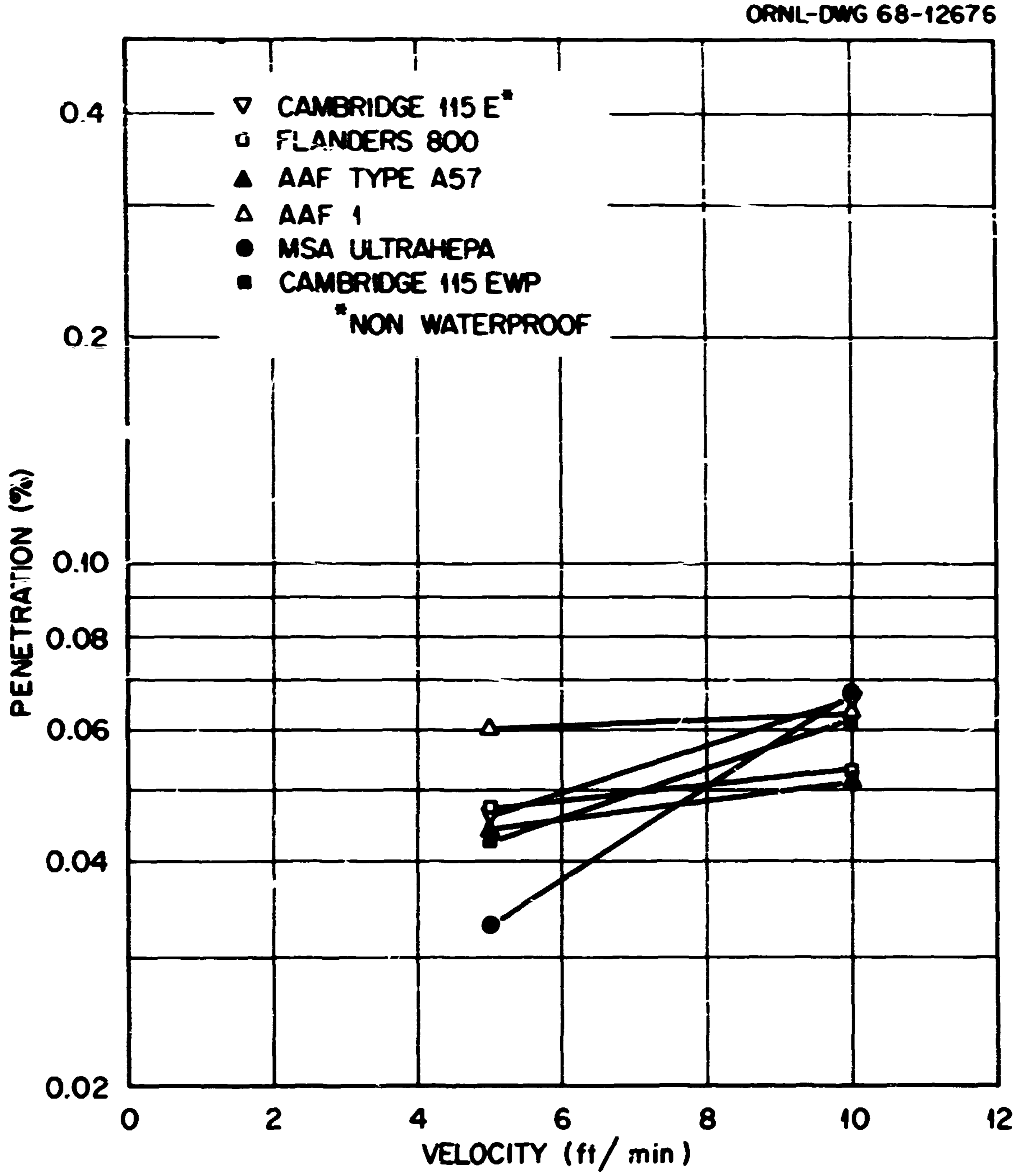

Fig. 7. Filter Efficiencies - Dry Stainless Steel-UO_2 Aerosol at 60°C.

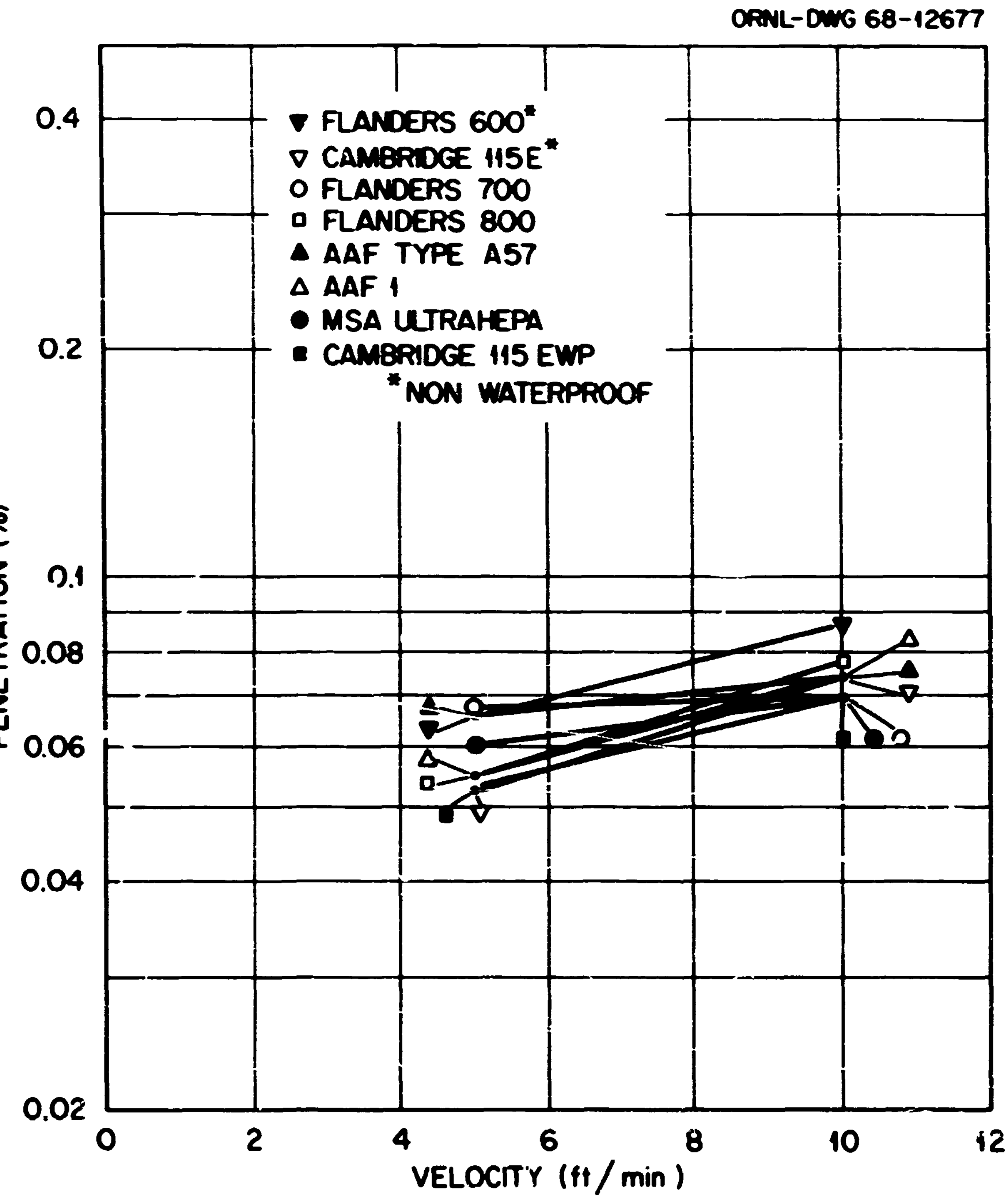

Fig. 8. Filter Efficiencies - Wet Stainless Steel-UO_2 Aerosol at 60°C.

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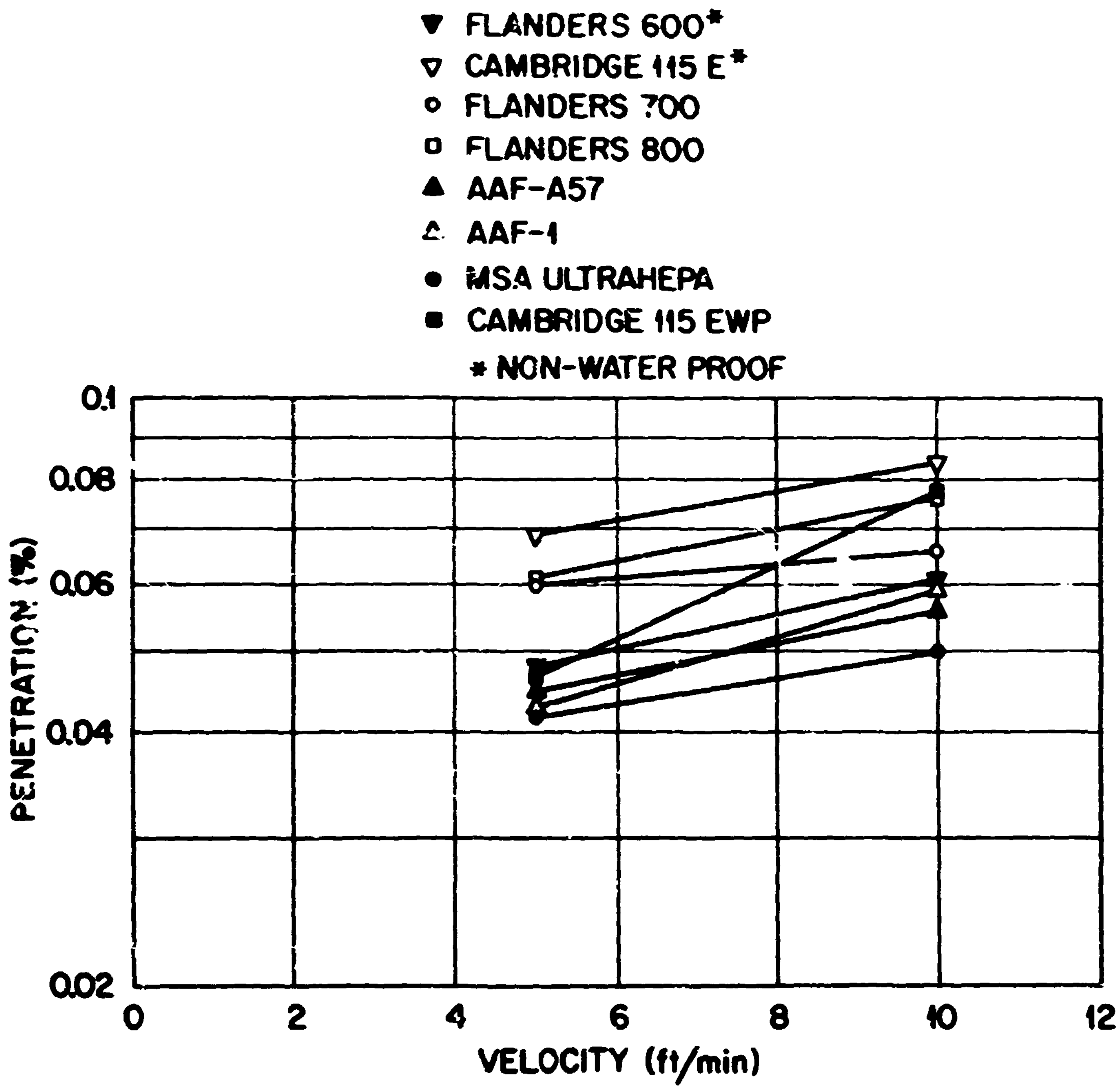

Fig. 9. Filter Efficiencies - Dry Stainless Steel-UO_2 Aerosol at 100°C.

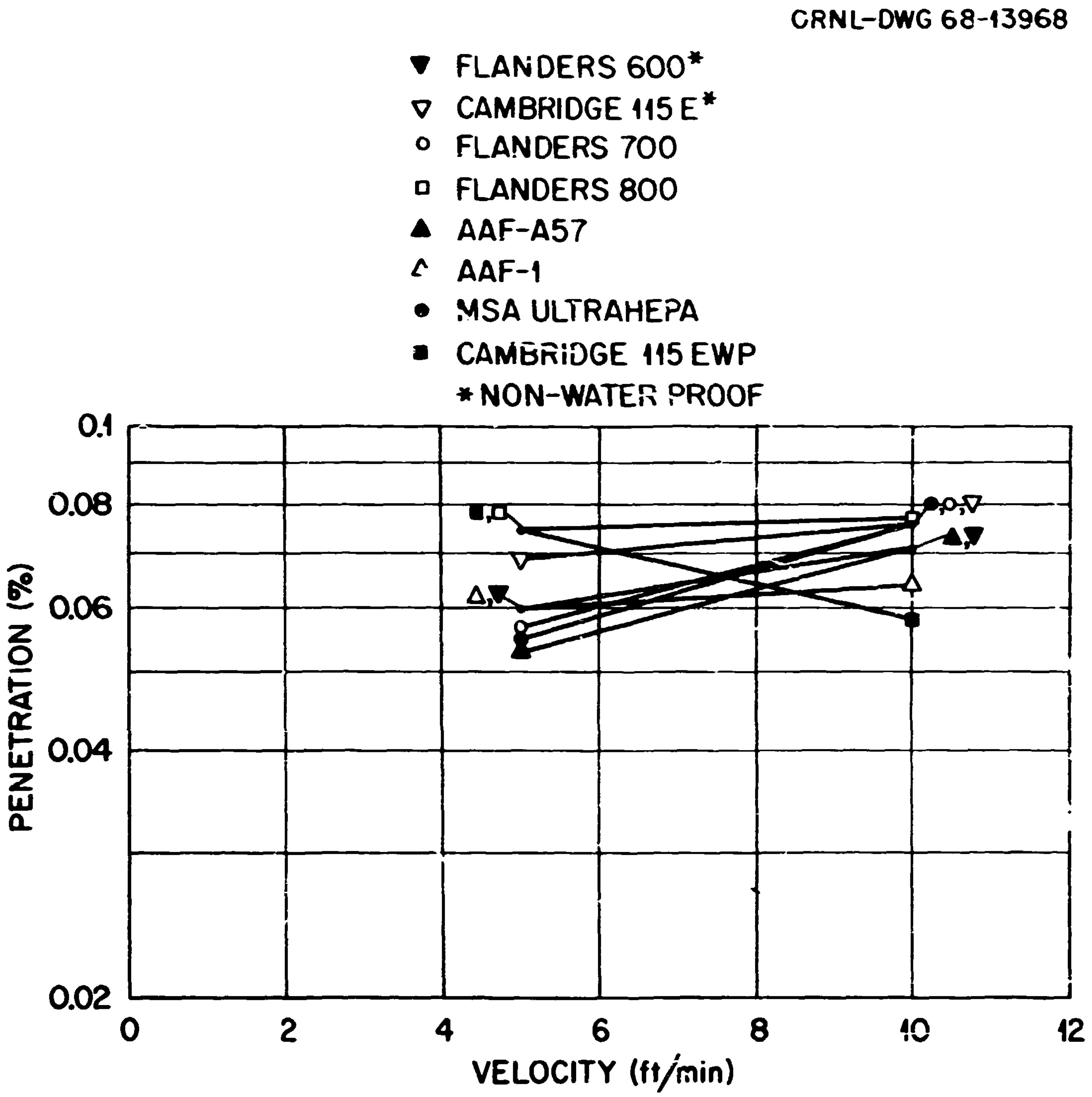

Fig. 10. Filter Efficiencies - Wet Stainless Steel-UO_2 Aerosol at 100°C.

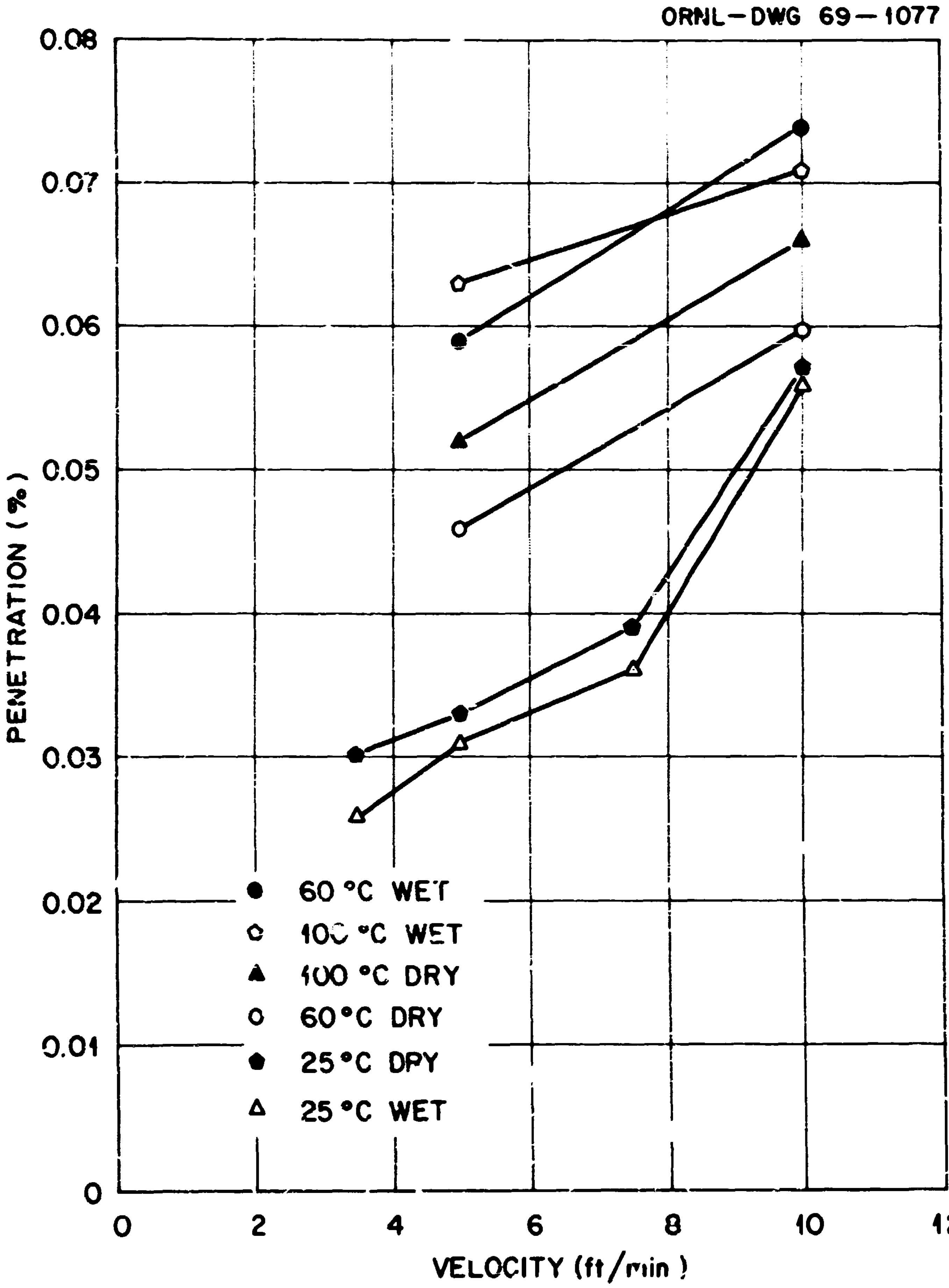

Fig. 11. Summary of Filter Penetration Results.

Fig. 12. Dry Stainless Steel Oxide Aerosol.

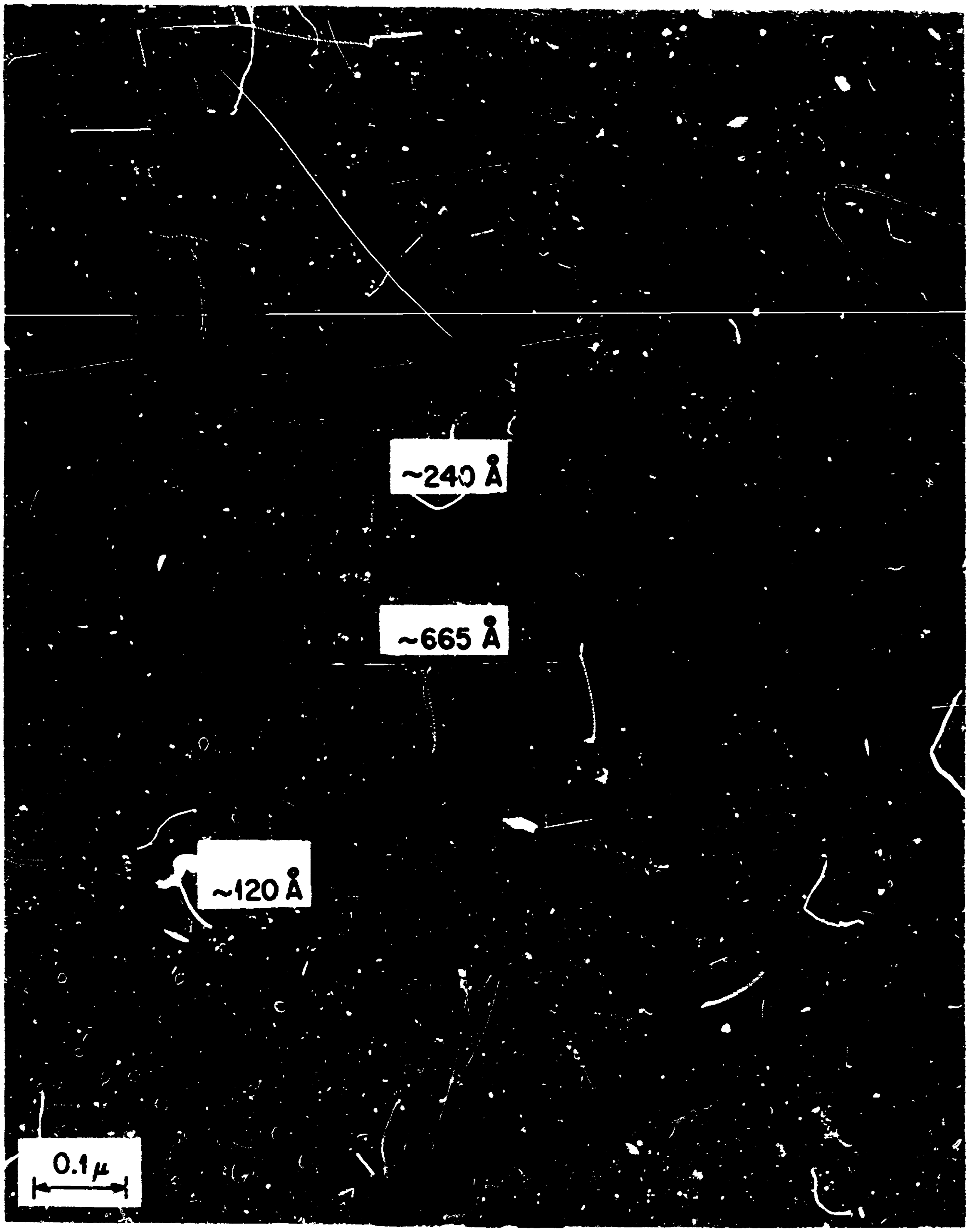

Fig. 13. Dry Stainless Steel-Oxide-UO_2 Agglomerate Particle.

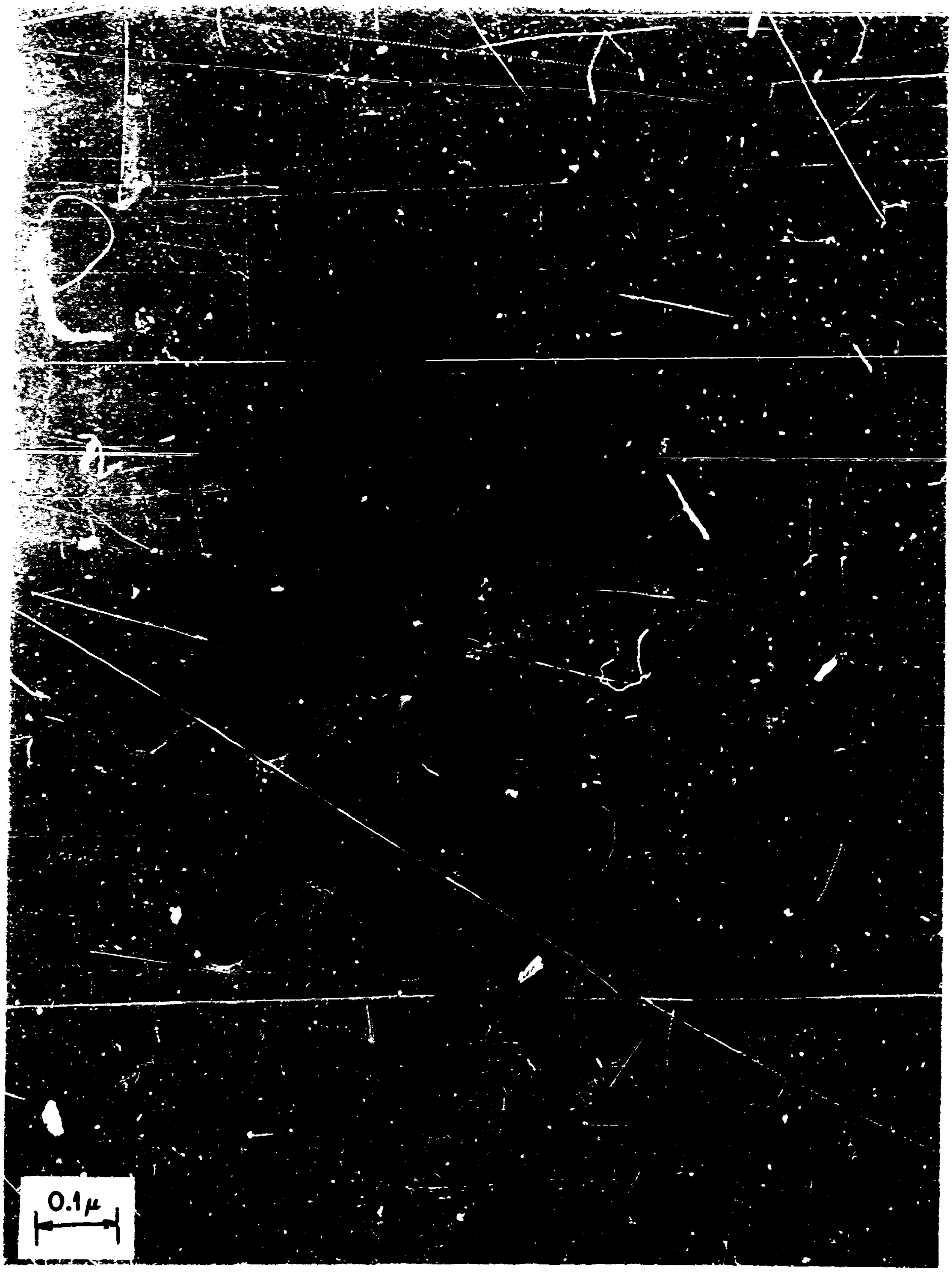

Fig. 14. Wet Stainless Steel-Oxide-UO_2 Agglomerate Particle.

Fig. 15. Dry Stainless Steel Oxide-ZrO_2 Agglomerate Particle.

Fig. 16. Wet UO_2 Agglomerate Particles.

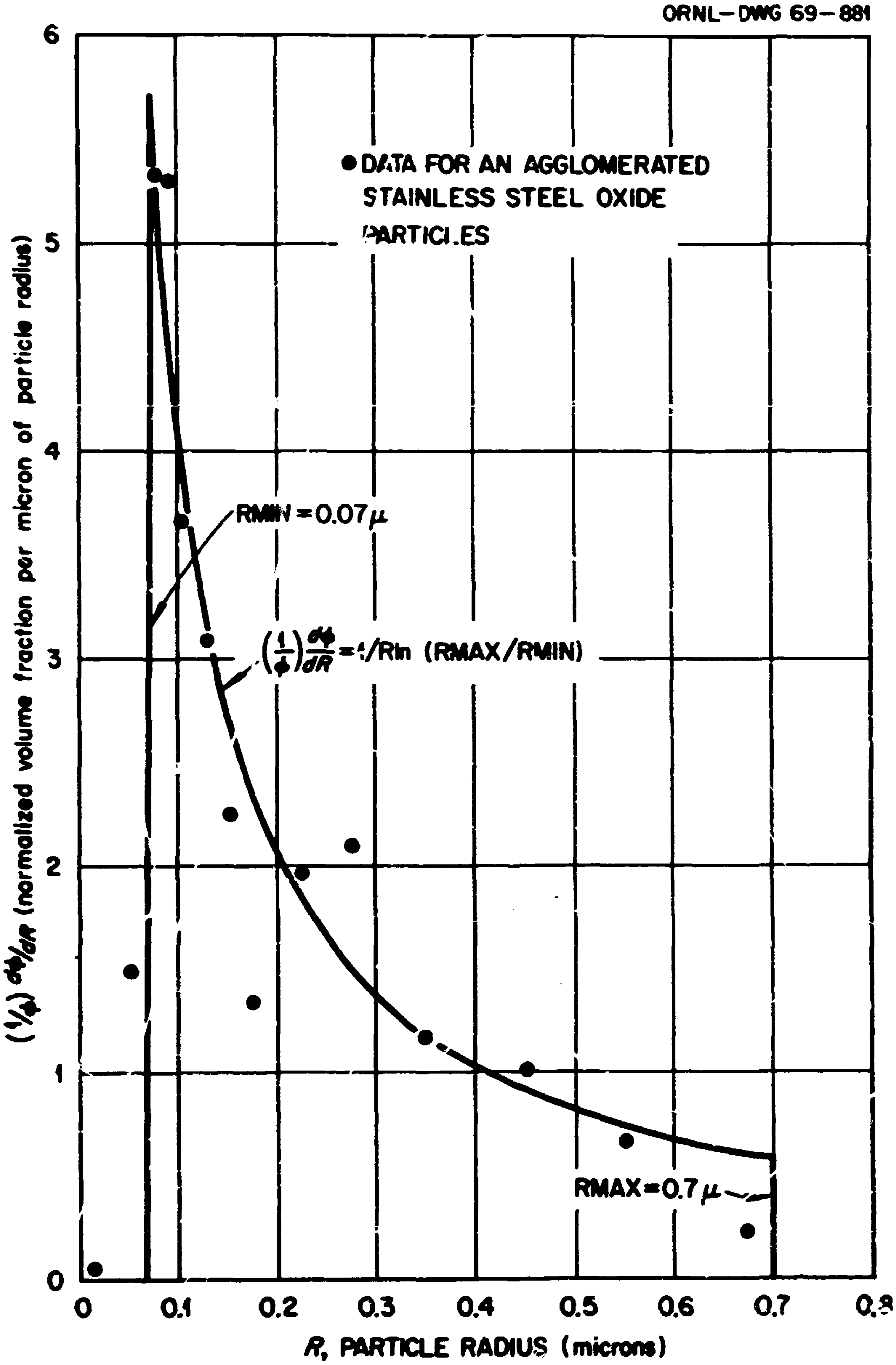

Fig. 17. Normalized Volume Fraction Distribution Function from Counting Instruments.

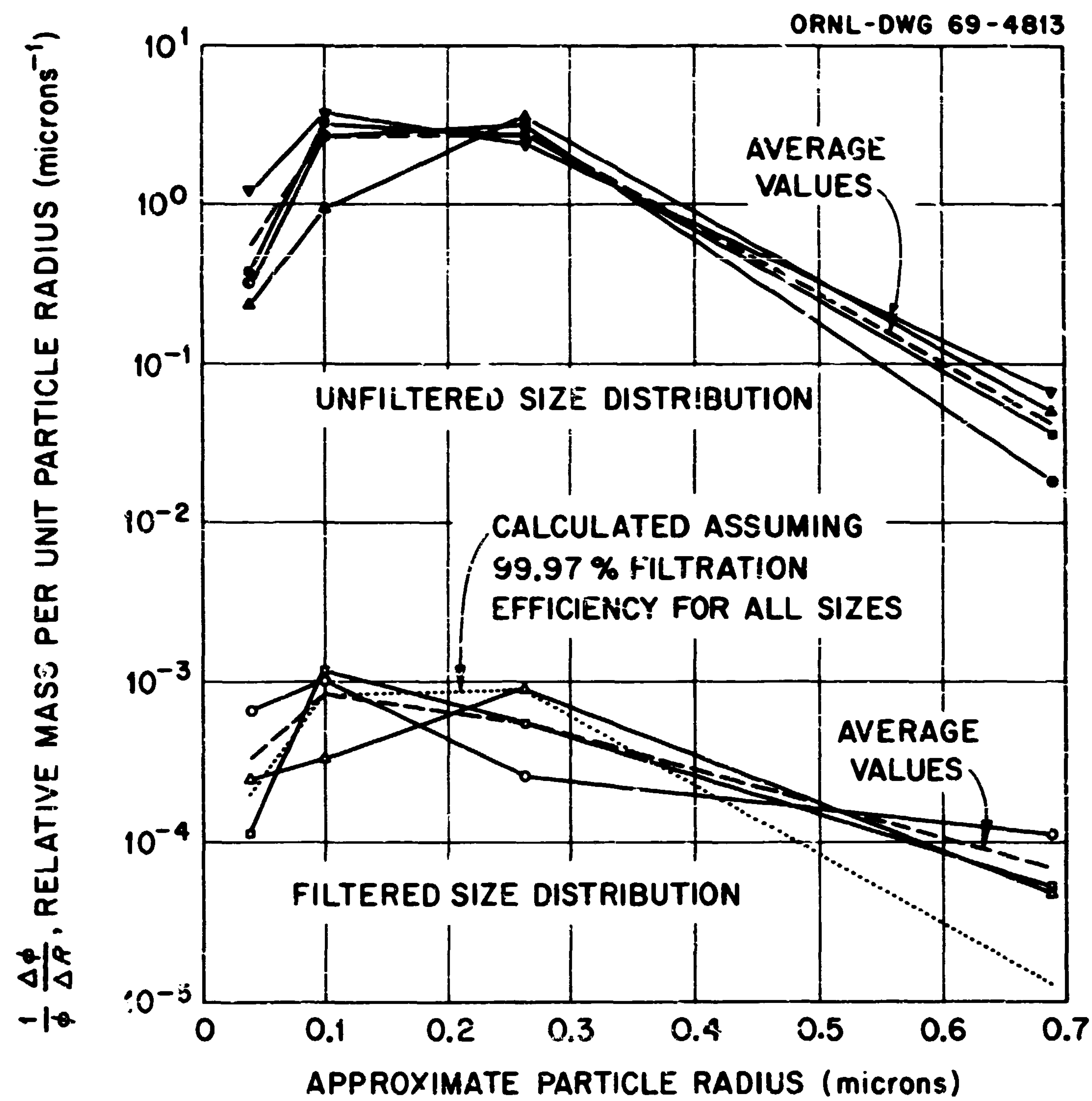

Fig. 18. Unfiltered and Filtered Particle Size Distribution from Low-Pressure Impactor Data.

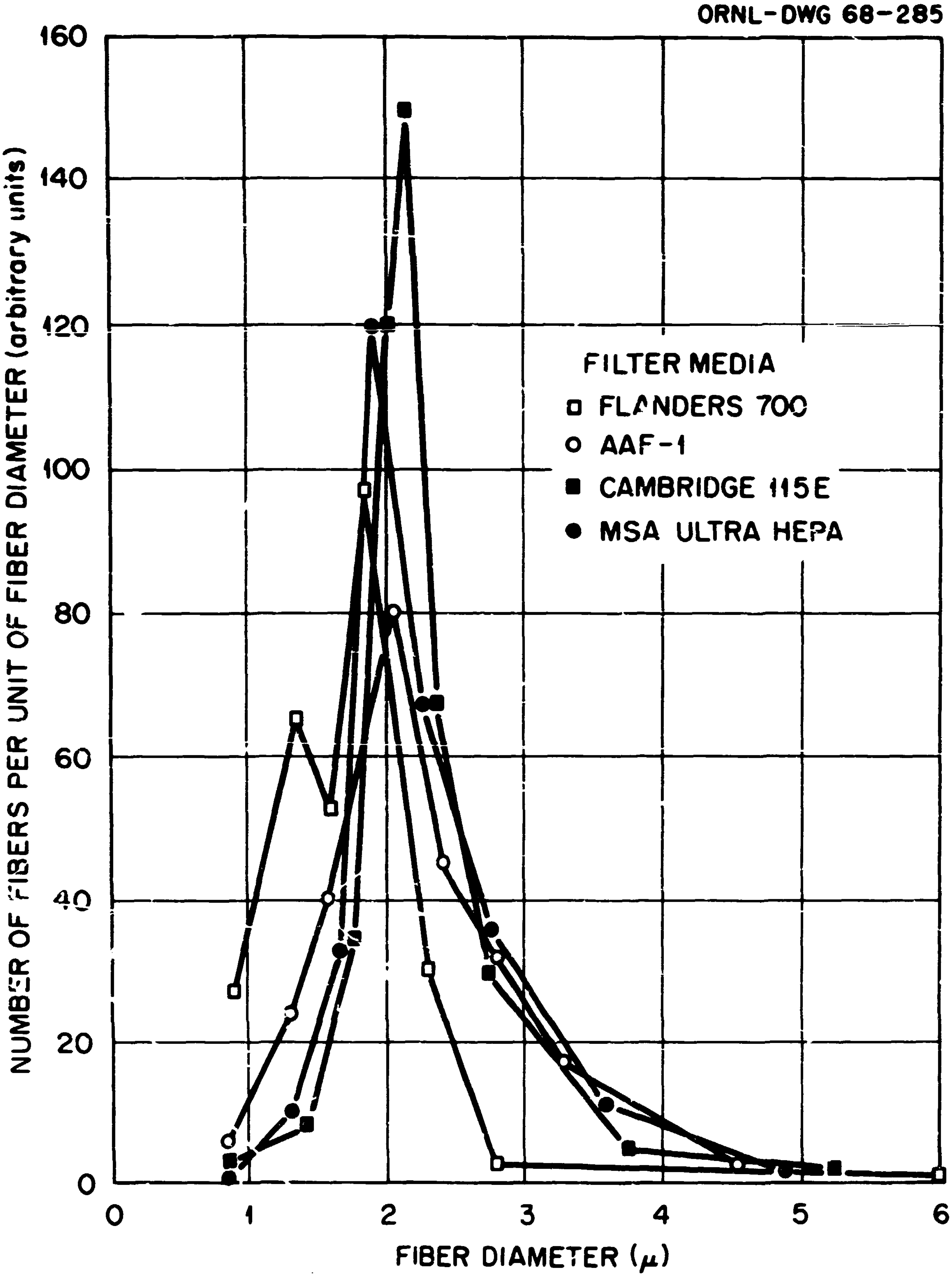

Fig. 19. Fiber Diameter Distribution Obtained by Optical Microscopy.

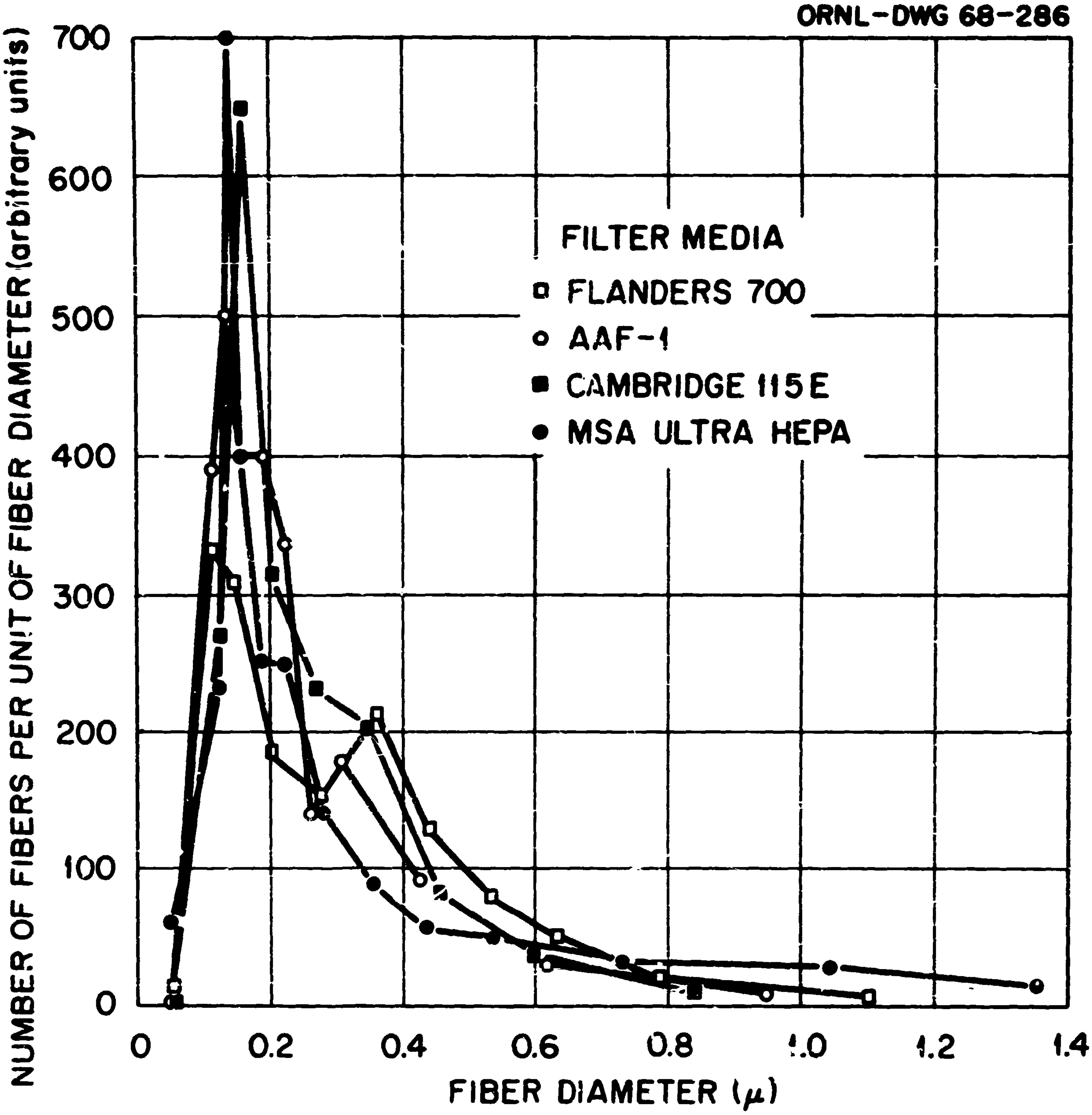

Fig. 20. Fiber Diameter Distribution Obtained by Electron Microscopy.

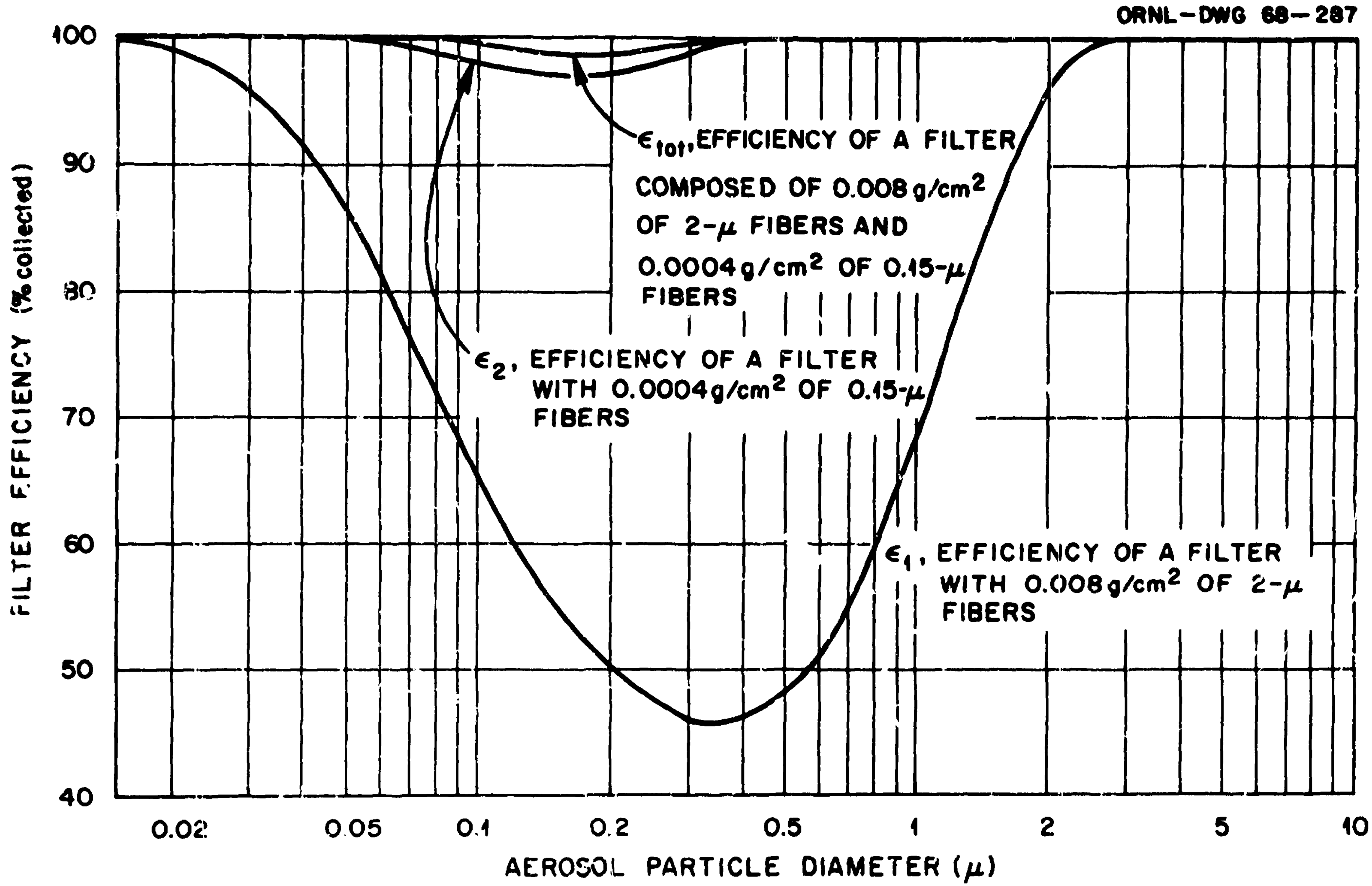

Fig. 21. Calculated Filter Efficiency vs Particle Size.

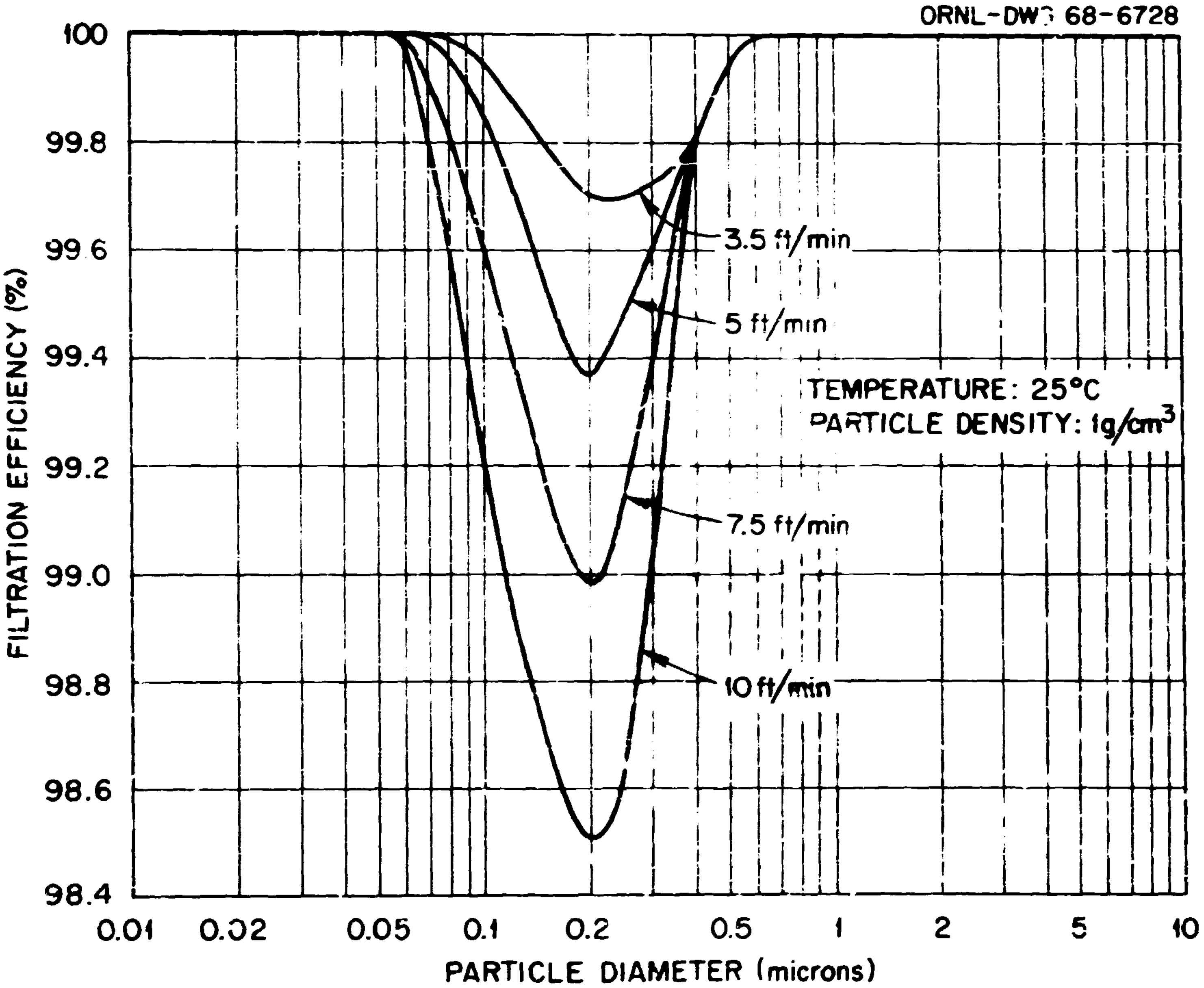

Fig. 22. Calculated Effect of Velocity on Filtration Efficiency.

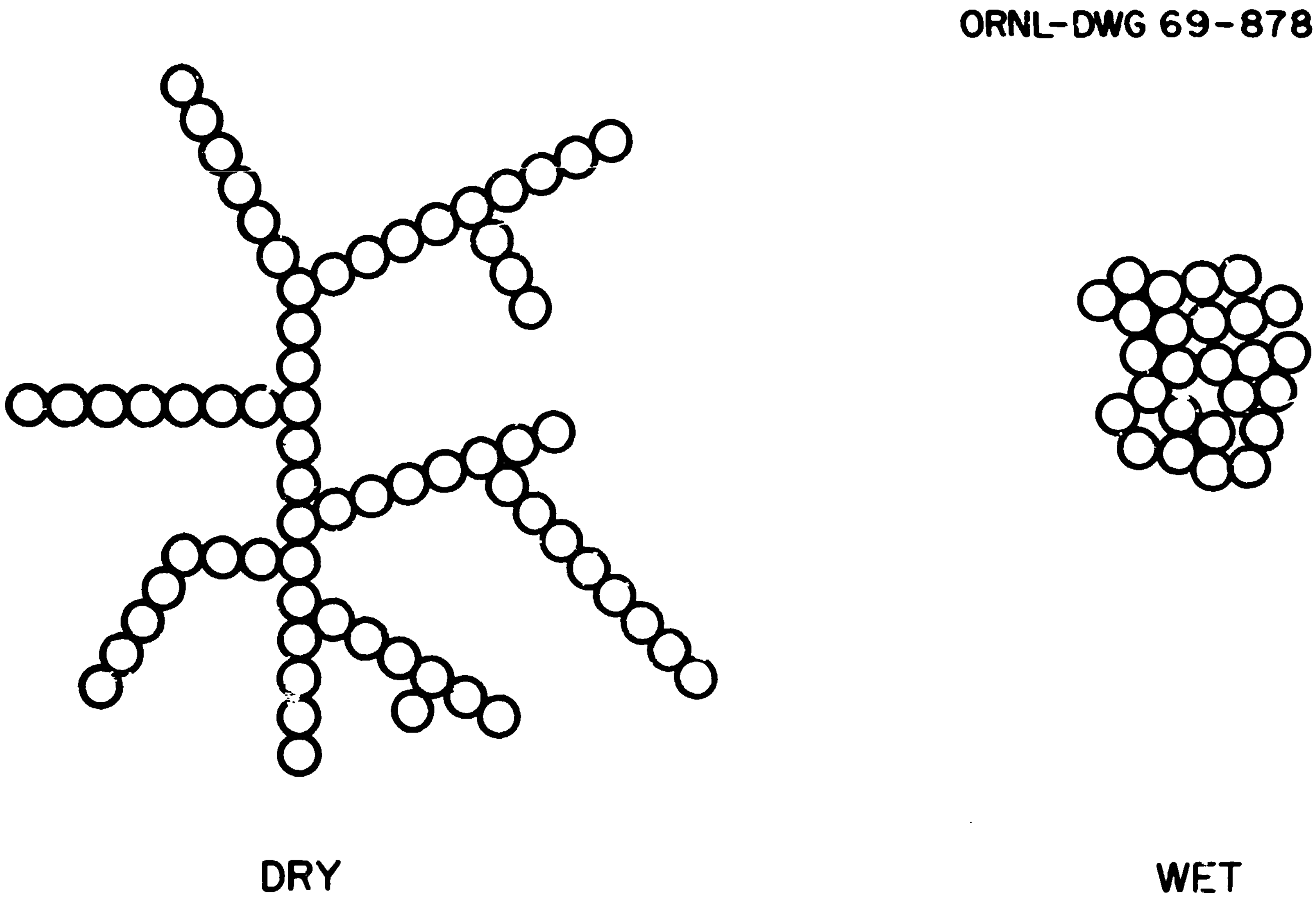

Fig. 23. Effect of Moisture on Agglomerate Shape.

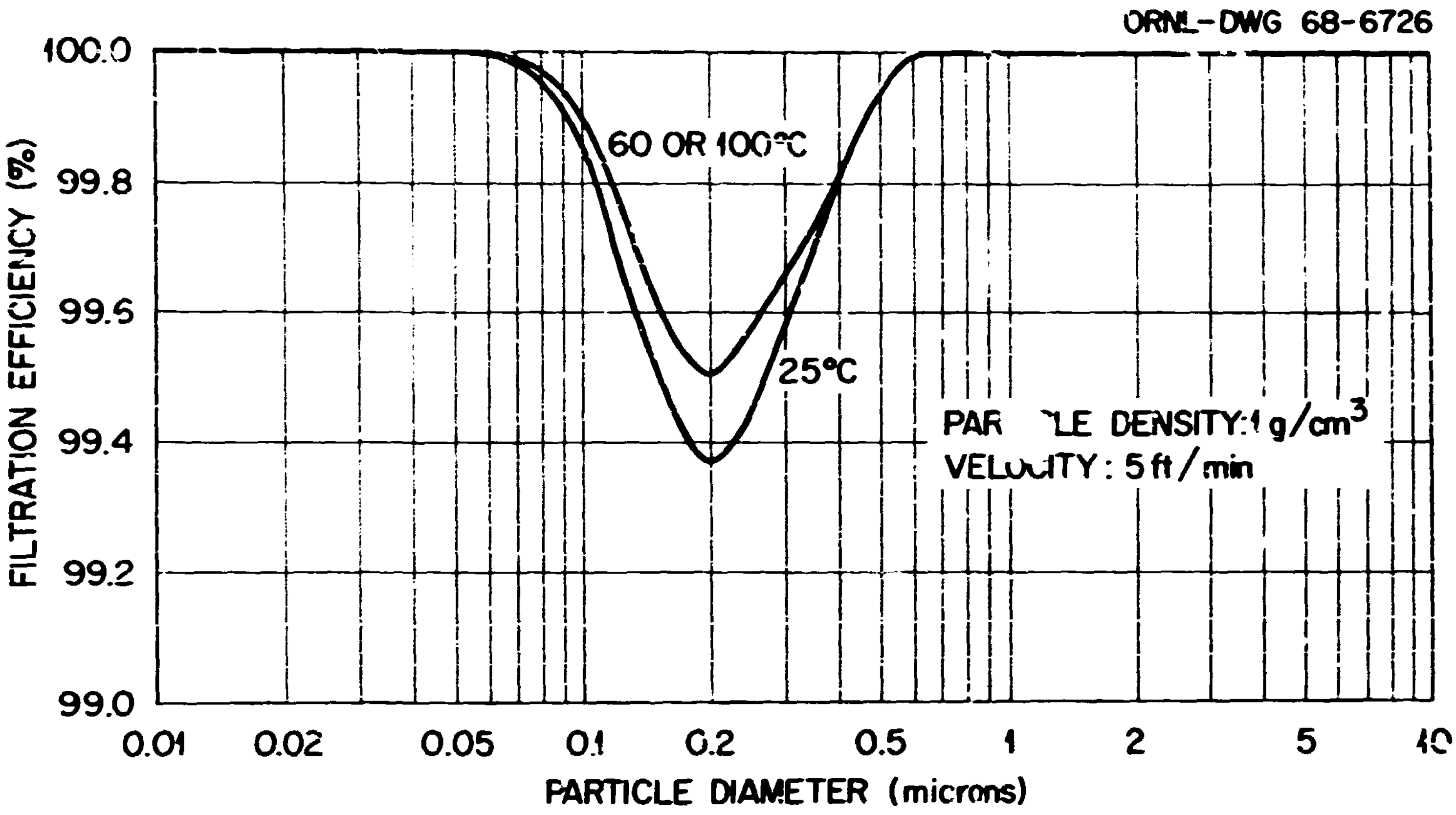

Fig. 24. Calculated Effect of Temperature on Filtration Efficiency.

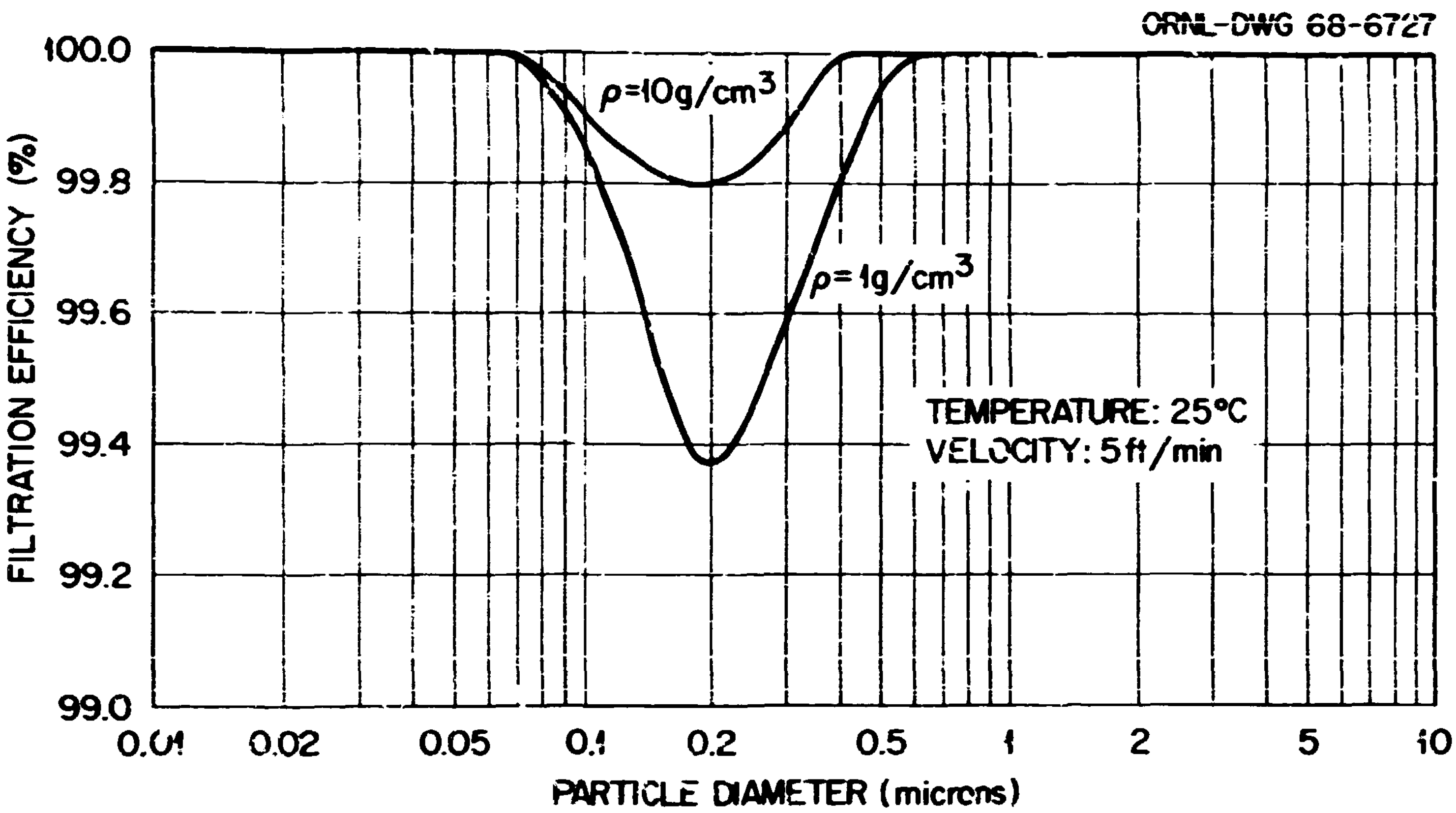

Fig. 25. Calculated Effect of Particle Density on Filtration Efficiency.

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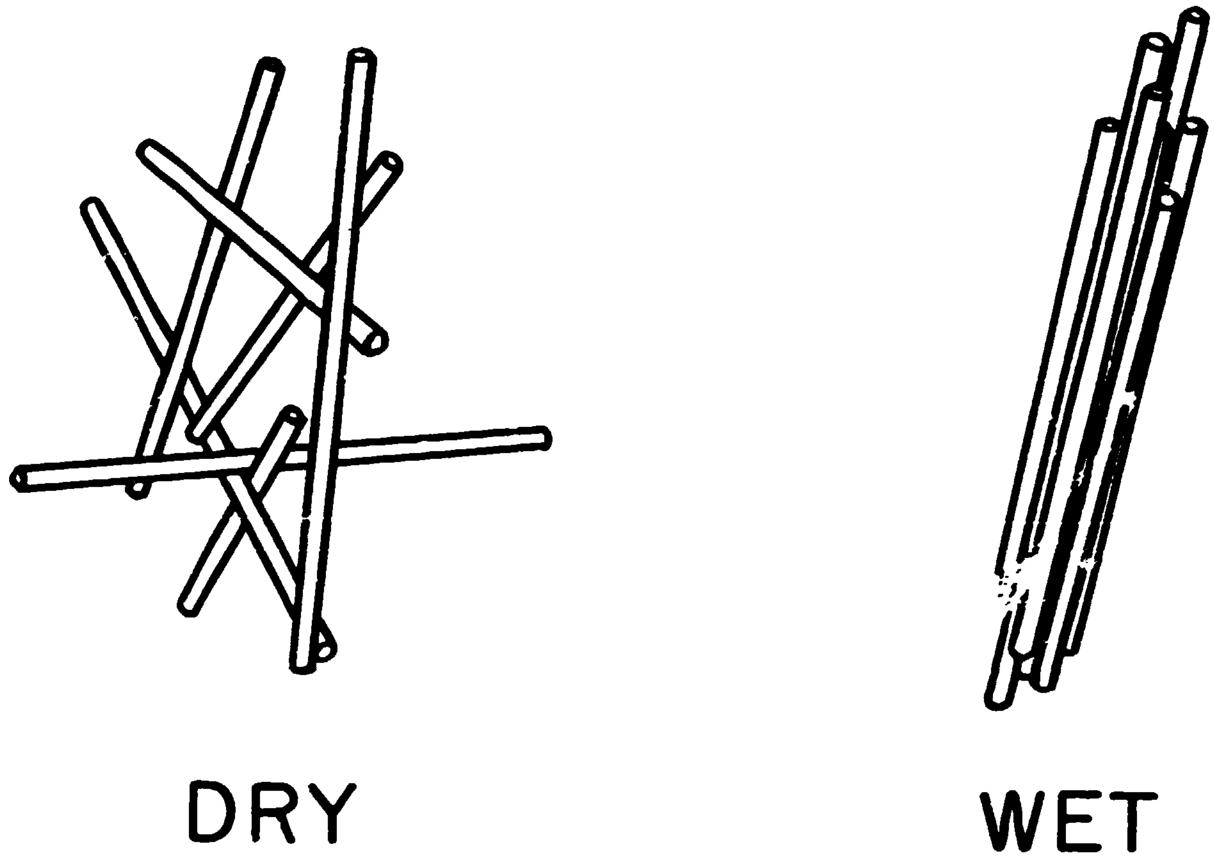

Fig. 26. Postulated Effect of Moisture on Fiber Size.

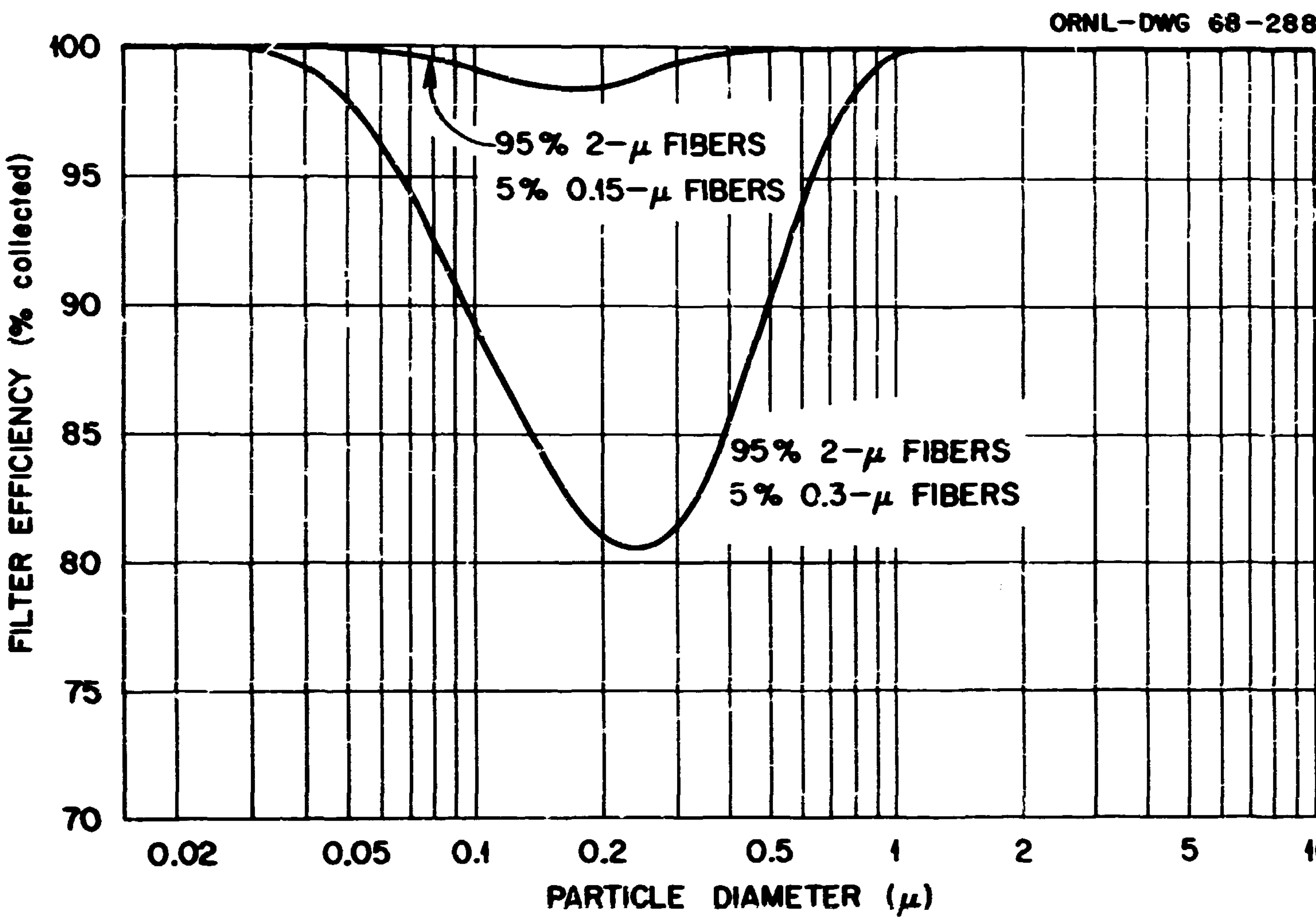

Fig. 27. Calculated Effect on Filtration Efficiency of Doubling the Diameter of Small (0.15 Micron) Fibers.

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