

Figure 8. Prefilter Holder as Used with HEPA Filter

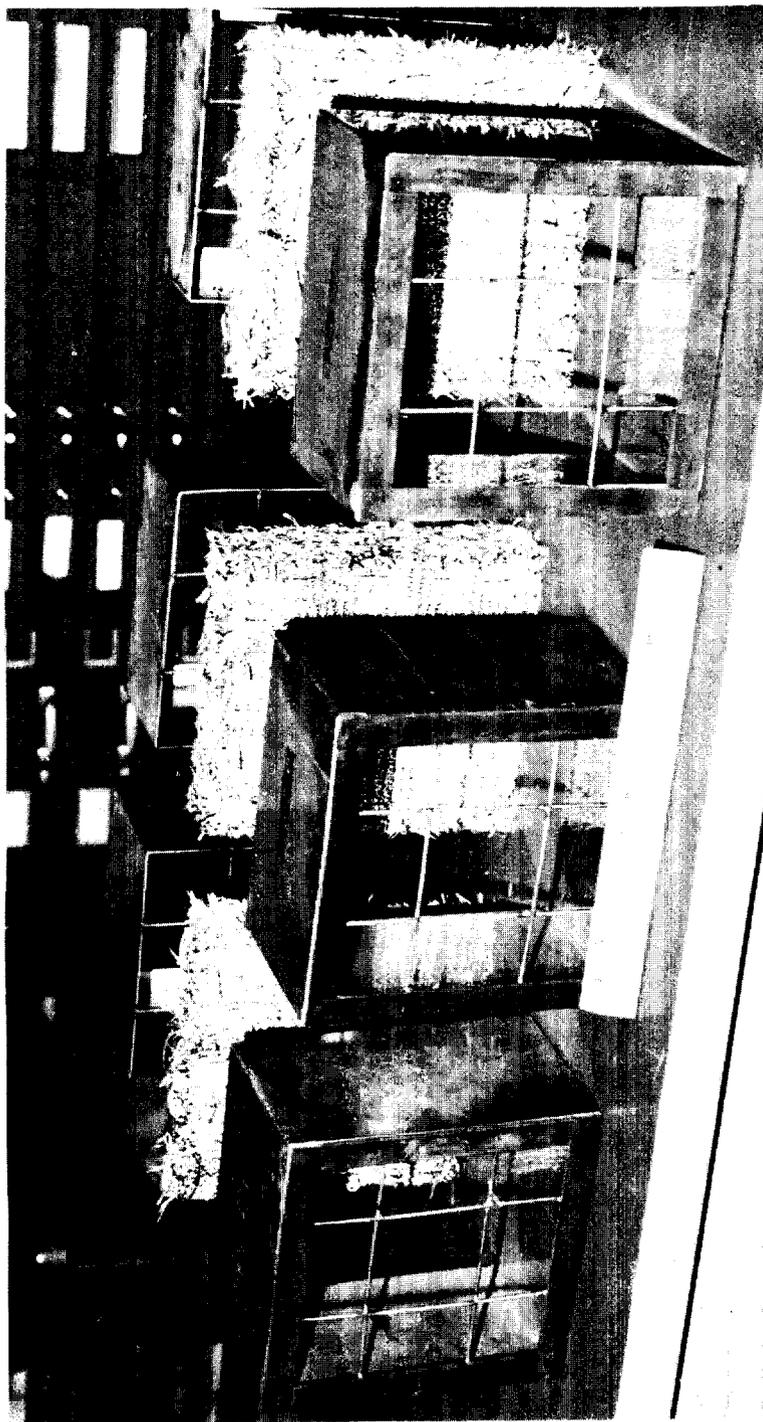


Figure 9. Prefilters Constructed of Stainless Steel
Frames and ACS Medium

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After thirty days of service, the prefilter had plugged. The prefilter assembly was removed, the medium was taken from its holding frame and washed in the glovebox enclosure using water. Although the medium was rinsed repetitively, it could not be cleaned completely. After draining, the medium was allowed to dry and then returned to service.

Later on during the test period, the medium compacted in the holder to the extent that an additional pad, one-inch thick, was added to supplement the volume originally occupied by the three-inch pad. The ACS medium and its holding frame were dismantled after five months of intermittent service under production conditions. Although the efficiency of this type of medium is good for coarse particulate and droplets (2), it does not lend itself well to cleaning under the washing procedure tested.

IV. Summary

For summary, work toward the objective of prolonging the life of HEPA filters is proceeding along several lines. We have shown that asbestos does impart chemical resistance to HEPA filters and a suitable substitute should be found since use of asbestos in HEPA filters is being phased out.

Also, chemically resistant materials are being investigated for the replacement of asbestos separators.

The use of cleanable prefilters to reduce loading of glovebox HEPA filters appears to not only prolong the life of the filters but also reduces the amount of plutonium discarded as nuclear waste.

In-service tests of new commercial HEPA filters thus far indicates that they have service lives at least equal to those that are normally in current use.

Acknowledgement

Technical assistance was provided by J. F. Capes, W. D. Gilbert, K. J. Grossaint, and C. D. Skaats.

References

- (1) W. T. Hense, U. S. Patent 3,594,993 (1971)
- (2) M. W. First, D. H. Leith, "ACS Entrainment Separator Performance for Small Droplet-Air-Steam Service," Report No. ACS-1 (1975), ACS Industries, Woonsocket RI

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DISCUSSION

BURCHSTED: We had an opportunity to see three experimental HEPA filters that had been stored in the attic of an AEC contractor's facility a few years ago. One had PVC separators, the others had unidentified plastic separators. The separators had softened and the filter pack of all three filters had sagged. I would suggest, therefore, that you view any filter with plastic separators with great caution.

GILBERT: About 16 or 17 years ago, filter purchasers, filter makers and filter papermakers, and fiber makers began to get together before each Air Cleaning Conference to talk about their mutual problems in an informal way. This nucleus of some 16 or 17 people made drafts of specifications which formed the basis for the filter that we know today. The deliberations of this informal group have been reported at each Air Cleaning Conference by Mr. Anderson, who is at the Naval Surface Weapons Center at Dahlgren, Virginia. By this conference, the original group of 16 or 17 has grown to about 125. This will be this year's report of the former informal filtration group that is now known as The Government-Industry Meeting of Filters, Media, and Media Testing.

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GOVERNMENT-INDUSTRY MEETING ON FILTERS, MEDIA, AND MEDIA TESTING

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Many of the accomplishments of the air filtration programs achieved thus far have been due to the efforts of an informed working group concerned with high efficiency filters. The existence of this group has now spanned eight air cleaning conferences and has shown successive growth and participation at each one. From the original handful of participants and their open discussion mode of operation, the meeting has progressed to an invited audience with a prepared agenda and, at this meeting, selected technical papers. At this, the 15th conference, 110 persons comprised the assembled body; 79 from industry, 15 from government, 10 contract investigators and 6 international guests.

The most recent session of this working group was held this past Monday morning and was devoted to a series of discussions on subjects of current interest. This session, following the precedents of earlier meetings, related the operating requirements placed on the high efficiency filter and the capacity of industry to meet them. To this end, the collected talents of the assembled body were unified toward the solution of the problems of the particulate filter, its components and methods of test. Representatives of all facets of the industrial complex were present, from the basic fiber suppliers, through the media producers, and finally to the filter unit fabricators. Research organizations from R&D government laboratories and academic institutions contributed status reports on work currently underway. Users at various levels expressed their problems and actively participated in the discussions.

The following review of the discussions may seem to be an agglomeration of information and show little continuity of subject. It is my intent to review for you, in abstract form, the items of committee deliberation. The items will be addressed in order of their discussion. At the recent session, six separate subjects were discussed. These were:

Revised Standards for Testing HEPA Filter Media

At the 14th Air Cleaning conference in Sun Valley, it was proposed that existing standards for testing of HEPA filter media be reviewed and that a single standard procedure be adopted. It was reasoned that this action would result in uniform requirements, specified techniques, and standardized equipment for testing that would produce comparable results at the different sites. In order to achieve this goal, a committee task force was established. This committee has met a number of times over the past two years and has completed a summary document titled "Efficiency and Flow Resistance Testing of HEPA Filters and Filter Media". A draft of this document (dated 7-4-78) was distributed at the meeting for comment. Since it is proposed to issue the final document as an ANSI standard, the chairman established a deadline of September 30, for receipt of

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comments and/or proposed revisions. Final publication will be contingent on the receipt and resolution of these comments and revisions.

A second action item from the Sun Valley conference was the determination of the need for a reference laboratory for purpose of consultation and calibration verification. The task force contacted filter paper manufacturers, filter fabricators, and users of these products and determined that the need for a reference laboratory was well established and would be invaluable for the correlation of tests results. In addition, the task force investigated a number of potential candidates for this role and conducted a technical/political/fiscal comparison of the proposed groups. Based on this study, it was recommended to the assembled body that a reference laboratory be established and that the Rockwell group at Rocky Flats be the selected site. A showing of hands signified the acceptance of both recommendations with only three dissenting votes out of the total assembly. Specific details of the mode of operation, as well as availability of services for the reference laboratory will be included in the proposed ANSI standard on filter and media testing. A point of contact at the Rocky Flats reference site is Mr. C. D. Skaats, Rockwell International, P. O. Box 464, Golden Colorado 80401.

Size Characteristics of DOP

Recent experience in the operation of the homogenous DOP generators used for filter media testing has raised the question as to the accuracy of the particle size and/or the size distribution of the generated aerosol.

Chuck Skaats of Rockwell International, Golden Colorado, reported on his work with the LAS-200 Laser Aerosol Spectrometer (PMS Co. of Bolder, Colorado) and its use for particle size measurement and distribution. Using this device, Skaats was able to draw some conclusions about the operation of the generating system:

1. The size distribution of the generated test aerosol was dependent on the system physical parameters. Pot temperature and variations in this temperature were critical in determining both size and distribution. Pre-conditioning of the air of spaces containing the instruments is essential for complete particle size control.
2. Disagreement exists between particle sizes measured by the photoelectric owl and size determined by the LAS-200.
3. Precise temperature measurement and controls necessary for stated particle size specifications will require improved instrumentation over what is currently being used. A complete copy of the Rockwell paper is attached to this report as Appendix A.

Confirmation of the Rockwell conclusions have been obtained by a more comprehensive investigation at the Harvard Air Cleaning Laboratory (HACL). William Hinds (HACL) reported to the group on their measurements of particle size and size distribution of the "hot DOP" aerosol. Using a PMS model ASASP-300 laser photometer

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for their studies, they compared particle sizes of various aerosols as determined by the photoelectric owl and the laser photometers. The LAS-200 photometer of Rockwell was also compared against the same aerosols. In addition, Hinds related his results with some theoretical work of Sinclair and reached the following conclusions:

1. The geometric standard deviation (GSD) of the Q-127 generator is about 1.15. This is sufficiently large to affect the interpretation of data from tests using the monodispersed aerosol.
2. The disagreement between the particle size measured by the photoelectric owl and the laser photometer is the result of aerosol polydispersity.
3. Variations in GSD's are believed to be the cause of interlab variability in penetration measurements.

Accepting these conclusions as factual, Hinds made a series of recommendations that included several options; these options ranged from changing the overall test size to conform more closely with the maximum particle size to using current equipment and technology and applying a correction factor based on a known GSD of a reference source. A complete copy of the Harvard paper is attached to this report as Appendix B.

Several comments on the two preceding papers are in order. It is acknowledged that particle sizes determined by the two instruments (owl vs. laser) are significantly different. However, it has not been demonstrated that this is a real phenomenon rather than a function of the calibration of the laser photometer. Such areas as refractive index differentials, types of median diameters determined and/or reported, physical calibration assumptions, etc. should all be considered before one assumes which device gives the more representative diameters. Further evaluations of interlab variability should be conducted before one concludes that GSD differential is the reason for experimental data variations. Parameters of flow, concentration, technique, leakage, etc. all must be considered and evaluated as part of the determination. It is expected that some on-going work at the reference lab and at Harvard will clarify some of these areas.

Glass Fiber Diameter Determinations

For many years the average diameter of microglass fibers has been determined by either an air permeability method (NRL) or a liquid drain time method (Williams Freeness). In general, both techniques gave comparable results at the larger microfiber diameters; deviations had been noted at the lower diameters especially those below 0.6 microns. In addition, neither technique supplied specific information on fiber diameter distributions. Cliff Cain, Johns-Manville, Denver, Colorado, presented a paper on their recent efforts to modify these existing techniques to make the test more responsive to both fiber diameter and diameter distribution. Using gas surface area measurements (BET) as the most accurate and reproducible data source, Cain compared the air permeability, liquid drain concepts as proposed by the NRL, Williams, Darcy,

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Kozeny-Carman, and Davies theoretical equations. From this work, we concluded that:

1. The porosity of the fiber mat must be accounted for in determining the volume-surface average fiber diameter using fluid flow techniques (air permeability or liquid drain).

2. Neither the NRL nor the Williams techniques adequately accounts for the variation in fiber diameter distribution inherent in production fiber samples.

3. Evaluations using the Davies modification of the Darcy equation does account for the variation of fiber diameter distributions.

4. The physical constants (Frazier Number) in the Davies equation can be correlated graphically with the BET derived volume-surface average fiber diameter.

Based on the results of the study, JM has derived a modified Williams Freeness vs. average fiber diameter calibration curve and uses this as a means of production fiber sizing. This has resulted in a more precise determination of the average fiber diameter and a better tool for quality control of the fiber production process. Test procedures are described in JM document FG-436-209. Sales literature (FF-91B 10-77) has been modified to reflect the code change-fiber diameter relationship in the standard fiber sizes (codes 100-112).

A complete copy of the JM paper is attached to this report as Appendix C.

QPL and Mil-Spec Status

George Smith, Chemical Systems Laboratory, Aberdeen Proving Ground reported on the status of the Qualified Products Listing for filter units currently managed by the Army. A listing (dated 23 December 1977) has been issued listing all manufacturers that have qualified their products in the various sizes and/or configurations. Copies of this listing were distributed at the meeting. A new QPL program will be conducted in FY 79 to supplement the existing lists with the new qualifiers. It is anticipated that the announcement for this FY 79 effort will be issued in November 1978, with testing to commence about the first of the following year.

Mil-Spec 51068, Revision E, on filter units is out for review and expected to be issued soon. Mil-Spec 51079, Revision C, on filter media is likewise out for review and expected to be issued if certain issue points can be resolved. Considerable debate was conducted on the specific issue of having two grades of filter media, with a color coding distinguishing between the two. Users questioned the technology gain/cost savings stated by the Army as the reason for the two media. Additional discussions were addressed to the length of time required for specification issue and the lack of response of the Army to the DOE input to the media specification. The committee chairman will work with Aberdeen and the filter media

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manufacturers in an attempt to resolve the remaining point of disagreement.

Filter Testing Program

A number of years ago, a "round-robin" filter test exchange was conducted between various laboratory, industry, and academic groups. The task force deliberations indicated that a renewal of this effort would be desirable and commissioned the reference laboratory, Rocky Flats, to initiate such a program. A variety of filter media has been collected from various media suppliers and a test group of filters assembled for the test program. These samples have been packaged in metal containers and forwarded to the various participants. Instructions with the samples requested that the test program be conducted using the standard operating procedure of the Mil-Spec 51079B and the resultant data forwarded to the reference laboratory. Analyses of the submitted data will be conducted by the reference laboratory and W. L. Anderson of the Naval Surface Weapons Center, Dahlgren, Virginia. Summarized data will be coded and forwarded to all participants.

Certification Program

Discussions at the 14th Air Cleaning conference expressed the need for a recognized certification program for in-house test personnel. Mel First (Harvard) reported to the group on his efforts to achieve this goal. A preliminary draft of proposed criteria for qualification was prepared and the Society of Mechanical Engineers was approached for possible endorsement and inclusion with their other certification programs, a rejection by this group has been received. The society's position that this special area of expertise was not in accordance with their overall policy of certification needs required another approach. After further deliberations, it was concluded that an alternate method would be to incorporate the program in an ANSI standard format. Following this approach, a draft of the standards for qualification has been prepared and distributed. Dr. First requested that comments, additions, and/or revisions be submitted by September 30, so that further actions can be expedited.

In conclusion, it should be re-emphasized that this informal working group, with its diversified representation, provides a means for a comprehensive and expedient solution to the problems of the filtration industry. The total effort has proven invaluable because it permits the surfacing and exposure of problems that might otherwise be lost in the quagmire of bureaucracy and management. The meetings are intended to be and actually are, a working level distribution of data and expertise as well as a progress report of ongoing projects in the particle filtration areas. To this end, we feel that we have been successful and future sessions are contemplated.

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Appendix A

A STUDY OF DIOCTYL PHTHALATE PARTICLES (DOP) GENERATED IN PENETROMETERS AND THE DEVICES USED CURRENTLY TO MEASURE THEIR SIZE

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The present method of measuring the 0.3 micrometer particle has been of concern to many users of Monodispersed DOP Penetrometers. Verification of the 0.3 micrometer is totally dependent upon the photoelectric particle-size meters (owls)* and the capabilities of the individual calibrating the owls and associated electronics.

Results from research work and from the study of the size of particles generated by a Monodispersed DOP Penetrometer indicate that what has been previously considered factual is now questionable.

In early 1977, the Test Facility at Rocky Flats purchased a specially designed Laser Aerosol Spectrometer (LAS 200) capable of measuring particle sizes in sixteen (16) separate channels from 0.1 to 1 micrometer at intervals of 0.055 micrometers. The results of each channel are printed as total count or shown visually on an oscilloscope. Other features were designed into the LAS 200 including portability.

One question deals with the use of the Laser to calibrate a DOP Penetrometer. The Laser itself is calibrated with Dow Latex spheres. The refractive index of the spheres is 1.508, which is near the refractive index of DOP, 1.485. We considered these refractory indexes close enough that the Laser would produce reliable results.

The Laser was first used to monitor particle size in a Q127 with no regard to concentration of DOP, a temperature of 156°C and with mechanical and visual "Owl" reading of 29°.

Verifying that the electronics and the mechanical "Owl" were calibrated according to standard procedures, samples of the DOP aerosol were taken from the mechanical "Owl" through the Laser at a flow of 0.2CFM with a printout every ten seconds. The results were also visible on an oscilloscope.

* "Owl" refers to the NRL Photoelectric particle-size meters. Visual means of measuring the intensity variation of each field in the mechanical analyzer.

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The printout and visual reading indicated that the particle size ranged from 0.3 to 0.6 micrometers. The curve displayed on the oscilloscope showed that the curve did not peak at 0.3 micrometers as was expected, but was very flat from 0.3 to 0.6 micrometers. Calculations from the printout indicated that only 10 to 15% of the total particles were in the 0.3 micrometer range. The concentration of DOP was approximately 130 micrograms per liter.

Operating Procedures 136-300-138, Edgewood Arsenal, recommends that the operating temperature should be 168 to 172°C to produce the 0.3 micrometers. We had previously found that these temperatures were too high to produce the concentration levels of 100 ± 10 micrograms. Therefore, temperatures were reduced until the level of concentration was obtained.

The first step was to reduce the heat, in 5° increments. Observing the reading on the oscilloscope during each 5° change, we could observe the change in the curve and regulate the heat to increase the percentage of the 0.3 micrometer range. At the same time, a definite drop in concentration levels was observed.

The temperature was continuously monitored by a Ralco digital thermometer which is certified to $\pm 1^\circ\text{C}$. At 145°C, the concentration level was approximately 75 micrograms per liter. Calculations indicated that 50% of the particles were in the 0.330 micrometer range. Eighty-four percent of the particles were less than 0.450 micrometers with a Geometric Standard Deviation of 1.36. Several attempts were made to reduce the temperature below 145°C. In each instance, no measurable aerosol was detected. For reasons undetermined, just prior to disappearance of the aerosol, the 0.3 micron particle size increased to 65% of the total count for approximately 20 seconds.

Each day for approximately two months, the Q127 was operated at the temperature of 145°C and measured by the LAS 200. After carefully studying the results, we concluded that the 145°C temperature would produce the best results.

During the entire study, no particular attention was given to the mechanical or visual "Owl". All measurements were solely dependent upon the LAS 200. Aligning the mechanical "Owl" and checking with the visual "Owl" and the particle size meter registering zero, we found the analyzer read 45°. Our original setting was 29° at 156°C. A joint test program was performed on March 23, 1978, at Harvard University on particle size versus the degree-of-angle (mechanical analyzer). The results of the latter study will be reported at the Government-Industry meeting.

Studies were also performed on the Q107 and Q76 Penetrometers. We found these units much easier to control; also, each unit produced a larger quantity of 0.3 micrometer particles for a concentration of 80 micrograms per liter.

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We found the Geometric Standard Deviation of the Q107 to be 1.34 with 84% of the particles less than 0.47 micrometers. The Geometric Standard Deviation of the Q76 was 1.28 with 84% of the particles less than .425 micrometers. Several attempts were made to reduce the Geometric Standard Deviation on the Q107 to 1.28. Some success has been realized with present temperature controllers.

A second study was performed on a polydispersed generator with a single 0.080" Alaski nozzle. The generator was installed in a chamber 13'x7'x8', and it produced a concentration level of 25 mg/m³. This particular chamber is used for respiratory fitting. The median of the observed particle sizes ranged from 0.383 to 0.400 micrometers with a Geometric Standard Deviation of 1.33 to 1.36. Previous information indicated the particle size was in the 0.7 micrometer range.

After a six-months study of the Q127, Q76 and Q107, the results have been analyzed and the following conclusions were reached:

1. Temperature measurement and control requires improved instrumentation.
2. Closer control of the ambient air and intake air of the Q76 and Q107 is required between 68 and 72°F.
3. Re-evaluate the mechanical analyzer and the 29° setting.
4. Consider changing the required 0.3 micron to 0.2. Our study indicated this is easier to produce than 0.3 micrometers and still permit the use of the 29° setting in the analyzer.

We definitely feel that other factors such as atmospheric pressure, humidity, and temperature of operating air, influence the generation of the 0.3 micron particle.

We also evaluated DOP from different manufacturers and found that some are easier to control at lower temperature and will still produce the 0.3 micron particles.

Should further study confirm the Harvard and Rocky Flats findings, the next question is what affect, if any, will this have on the media and filter industry.

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Appendix B SIZE DISTRIBUTION OF "HOT DOP" AEROSOL PRODUCED BY ATI Q-127 AEROSOL GENERATOR

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Abstract

Measurements of the particle size distribution of the "Hot DOP" aerosol produced by the ATI Q-127 aerosol generator at mechanical analyzer settings of 25°, 29°, 35°, 40°, and 45° were made using a high resolution optical counter, PMS Model ASASP-300. Size distributions were approximately log normal. At mechanical analyzer settings of 29° and 45° the generator produced an aerosol with CMD's of 0.23 and 0.30 μm , respectively, and GSD's of 1.15. These CMD's are 23% and 13% less than the sizes expected from Sinclair's³ curve of analyzer angle versus diameter for monodisperse aerosols. The differences between measured CMD's and expected sizes for the given analyzer settings are shown to be due to the polydispersity of the aerosol and the fact that polarization ratio is an extremely strong function of diameter (proportional to $d^{0.1}$).

Experimental

Measurements were made at Harvard School of Public Health of the size distribution of the aerosol droplets produced by our ATI Q-127 "Hot DOP" aerosol generator. Figure 1 shows the airflow system used to make particle size measurements. A dust-free dilution system was needed to reduce aerosol concentrations to a level that could be accommodated by the particle size measuring instruments used. Room air passed through a standard 1000 cfm HEPA filter to remove dust particles which might otherwise have confounded measurements of DOP concentration and size distribution. The filter was tested in place with cold DOP and found to have penetration less than 0.05%.

Downstream of the HEPA filter, DOP aerosol from the Q-127 was introduced through a tube at the duct centerline. The aerosol then passed a Stairmand disk which mixed the concentrated DOP with dust-free air to produce a uniform concentration within the duct. Approximately six duct diameters downstream of the Stairmand disk, aerosol samples were withdrawn through a sampling port flush with the inside of the duct wall. The remaining aerosol passed through a Venturi flowmeter and flow regulating damper to an exhaust fan.

The Q-127 generator was operated at the recommended air rate of 100 liters per minute. The flow of DOP to the air dilution system was controlled by the fraction of the total that was passed to waste through the chuck; the balance passed to the filtered air dilution system. By changing the volume rate at which DOP entered the dilution air system, changes in DOP concentration were effected.

Size measurements were made with a PMS Model ASASP-300 active scattering aerosol spectrometer probe connected to a DAS-32 data control system and a Wang 2200 minicomputer. Samples were drawn through the spectrometer with its own sampling pump. Because all

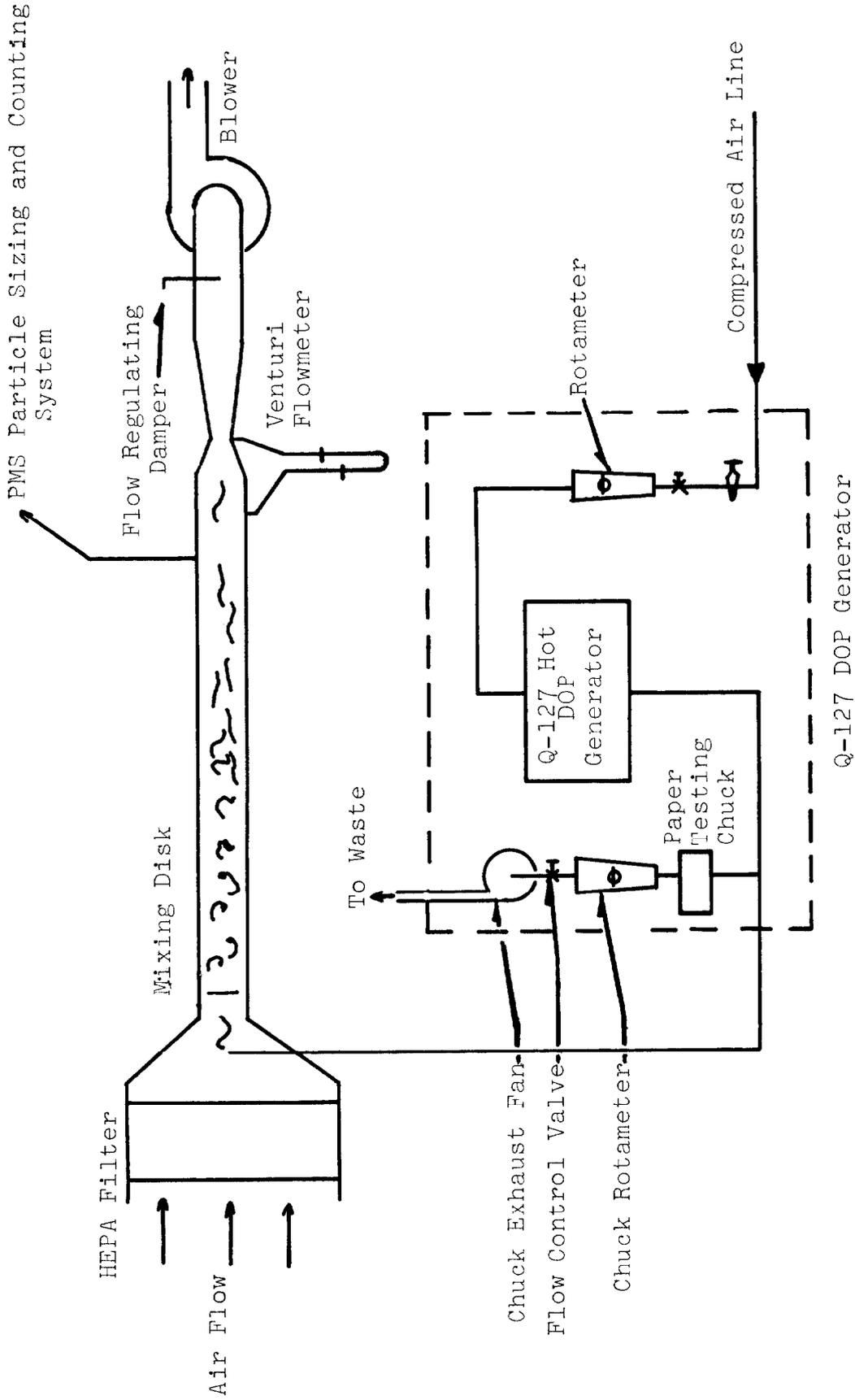


Fig. 1. Schematic drawing of "Hot DOP" air dilution system

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the DOP particles were submicrometer, no attempt was made to sample from the duct isokinetically. The relevant size ranges of the instrument are shown in Table 1. Because the spectrometer uses two independent instrument settings to cover the size range of interest, a check was made to ensure that these ranges overlapped properly. Results, shown in Figure 2, show good agreement in the overlapping region.

Immediately prior to sampling, background size distribution was measured and instrument size calibration verified using an aerosol of uniform diameter, 0.206 μm , polystyrene latex (PSL) spheres. The measured background particle counts are shown in Table 1. Sampling time for the background counts was 10 minutes but two minutes was used for the high concentration DOP measurement runs. The PSL spheres used for calibration were labeled 0.234 μm by Dow Chem. Co. but were found to be 0.206 μm by Porstendorfer and Heyder.¹ The light scattering instrument manufacturer claims that instrument accuracy is equal to plus or minus one channel width and the primary peak shown in Figure 3 is within this range.

Table 1. Ten minute background counts for test system.

PROBE A RANGE 3				PROBE A RANGE 2			
.15 TO .3 BY 0.010				.23 TO .605 BY 0.025			
I	MIN	MAX	COUNT	I	MIN	MAX	COUNT
1	0.150	0.160	20	1	0.230	0.255	5
2	0.160	0.170	2	2	0.255	0.280	2
3	0.170	0.180	4	3	0.280	0.305	0
4	0.180	0.190	2	4	0.305	0.330	1
5	0.190	0.200	2	5	0.330	0.355	0
6	0.200	0.210	3	6	0.355	0.380	1
7	0.210	0.220	3	7	0.380	0.405	0
8	0.220	0.230	1	8	0.405	0.430	0
9	0.230	0.240	1	9	0.430	0.455	0
10	0.240	0.250	0	10	0.455	0.480	0
11	0.250	0.260	1	11	0.480	0.505	0
12	0.260	0.270	1	12	0.505	0.530	0
13	0.270	0.280	1	13	0.530	0.555	0
14	0.280	0.290	0	14	0.555	0.580	0
15	0.290	0.300	2	15	0.580	0.605	0

Measurements were recorded in a format similar to Table 1 and results plotted on log probability graph paper as cumulative size distributions. Count median diameter and geometric standard deviation were obtained by eye fit of the best straight line through points between cumulative counts of 10 and 90 percent. The first set of DOP measurements was taken at an "OWL" analyzer angle of 29°, the "standard setting" for the Q-127 generator.⁴ Measurements were made at two concentrations differing by a factor of 6.4 to determine the significance of coincidence effects on particle size measurements. Results for a single measurement at each concentration are summarized in Table 2. Although the CMD's are within experimental error, the GSD is slightly higher at the higher concentration, consequently all subsequent measurements were made at a DOP aerosol concentration of 75 $\mu\text{g}/\text{m}^3$.

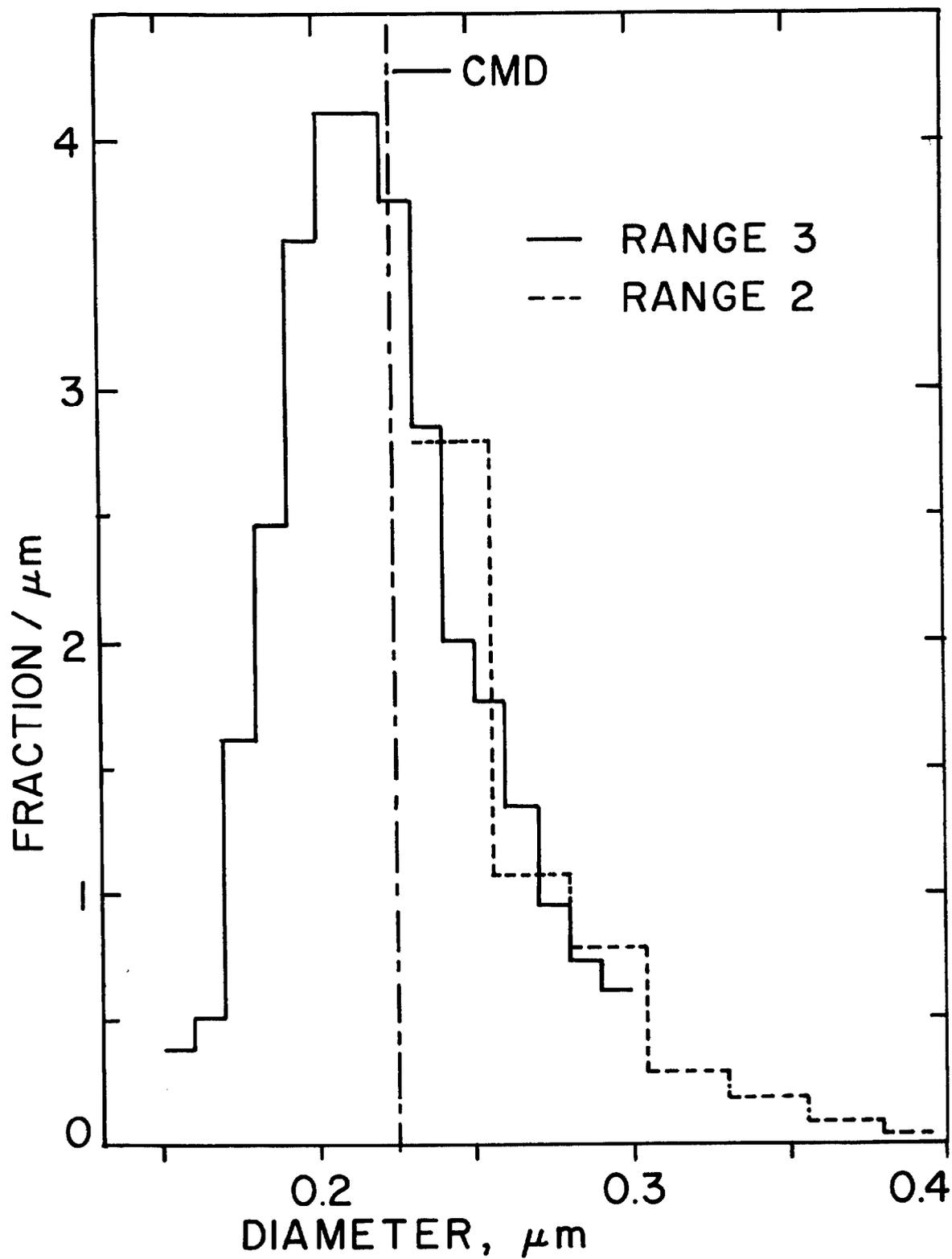


Fig. 2. Size distribution of "Hot DOP" showing overlap of PMS size ranges 2 and 3.

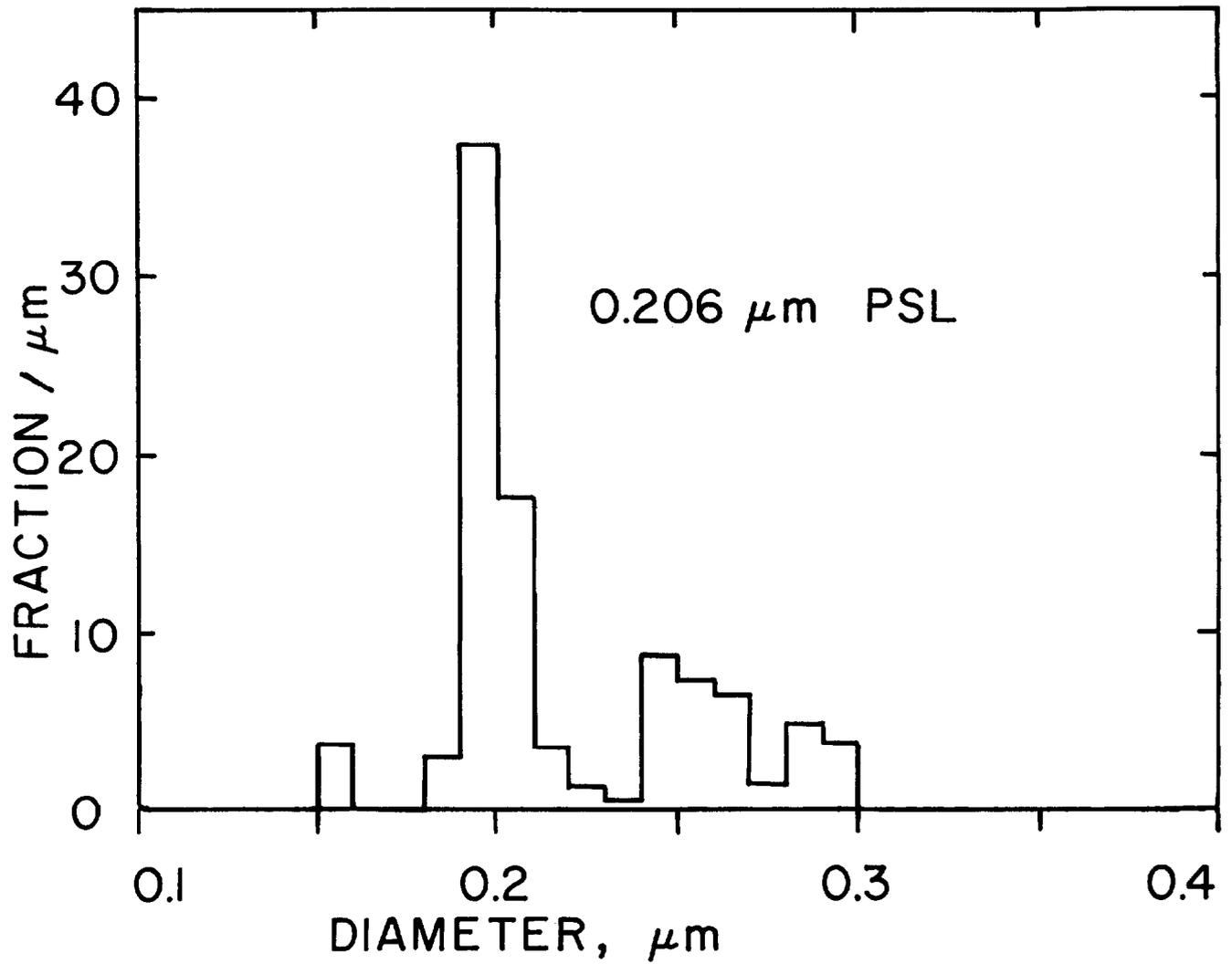


Fig. 3. Calibration of PMS with 0.206 μm PSL

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Table 2. Size distribution parameters measured by the PMS instrument at two aerosol concentrations for an analyzer setting of 29°.

<u>DOP Conc.</u>	<u>CMD</u>	<u>GSD</u>
75 $\mu\text{g}/\text{m}^3$	0.23 μm	1.18
480 $\mu\text{g}/\text{m}^3$	0.24	1.24

A second set of measurements was made to determine the size distribution for different generator operating conditions corresponding to OWL analyzer angles of 25°, 29°, 35°, 40°, and 45°. Results are shown for 29° and 45° in Figures 4 and 5, and all results are summarized in Table 3. As expected, particle size increased with analyzer angle setting and GSD remained constant for all analyzer angle settings. For analyzer angles of 29° and larger, a secondary peak was observed in the one or two smallest size channels. This secondary peak was not observed by Tillery² using a different PMS spectrometer with 0.01 μm channel width over the range of 0.08 to 0.23 μm . For the purpose of this analysis the small diameter secondary peak was considered to be an artifact of our instrument and was ignored in the size analysis. Figure 6 shows a comparison of Sinclair's curve of size vs. polarization ratio for monodisperse aerosols³ and the count median diameters we obtained at the five analyzer angle settings.

Table 3. Size distribution parameters measured by PMS instrument at six analyzer settings.

<u>Analyzer Setting</u>	<u>CMD</u>	<u>GSD</u>
25°	0.205	1.18
29°	0.232	1.15
35°	0.250	1.15
40°	0.265	1.15
45°	0.300	1.15

It was suggested that the difference between measured size and theoretically derived size of 0.3 μm might be related to a difference in index of refraction (IR) between PSL calibration particles and DOP particles produced by the Q-127. Table 4 shows IR values for PSL and for DOP derived from several sources: "New DOP" was removed from the shipping containers in which it was received, "Used DOP" was drained from the reservoir of the generator after a number of hours of operation, and "Aerosol DOP" was collected by electrostatic precipitation of the output aerosol of the Harvard Q-127. The differences in IR of the several DOP samples is considered to be insignificant with respect to the response of the PMS analyzer and the same is true for the differences in IR between DOP and PSL. Knollenberg⁷ has discussed the insensitivity of the PMS analyzer to changes in IR of aerosol particles.

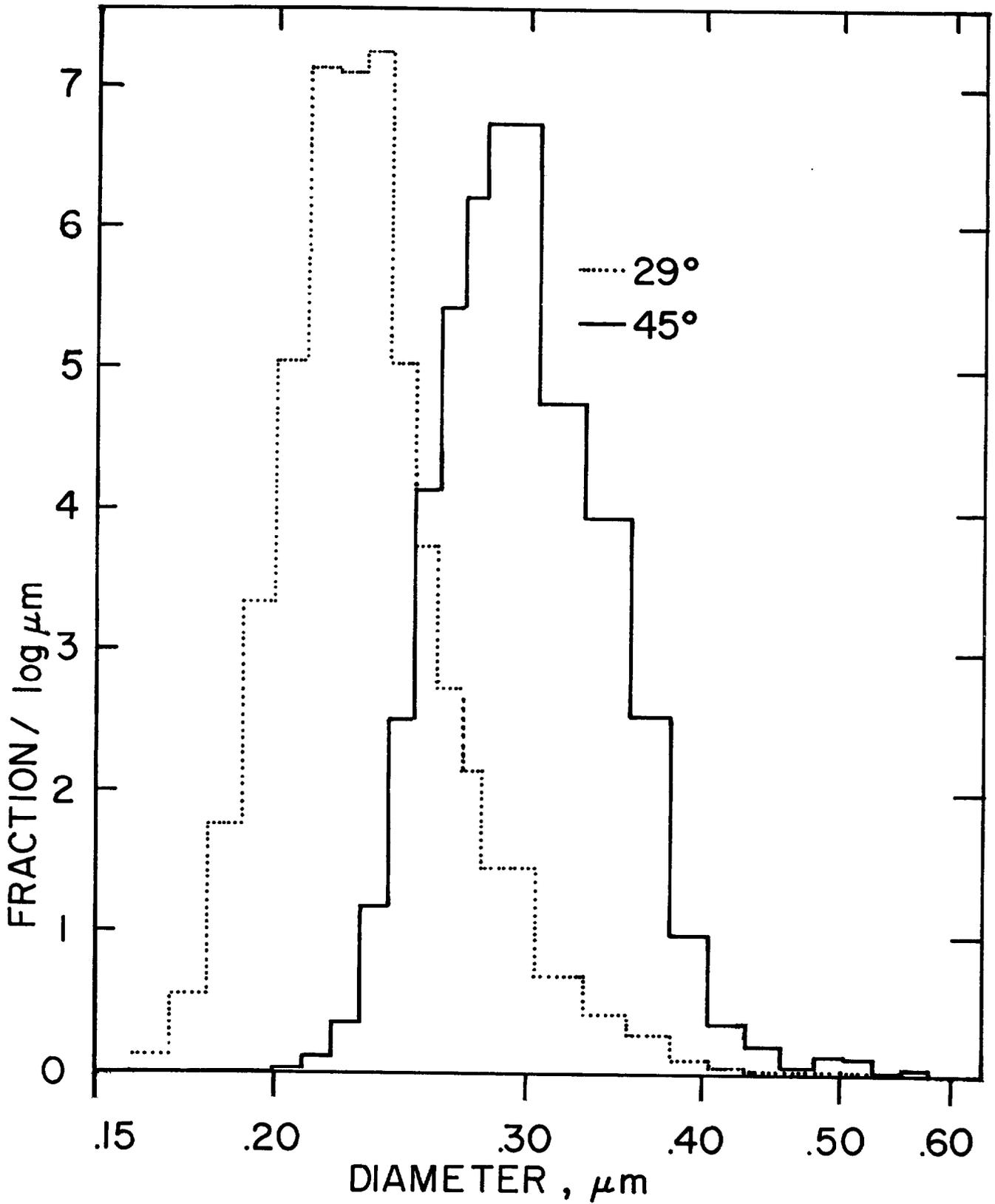


Fig. 4. Size distribution of DOP aerosol produced by Q-127 generator at analyzer settings of 29° and 45°.

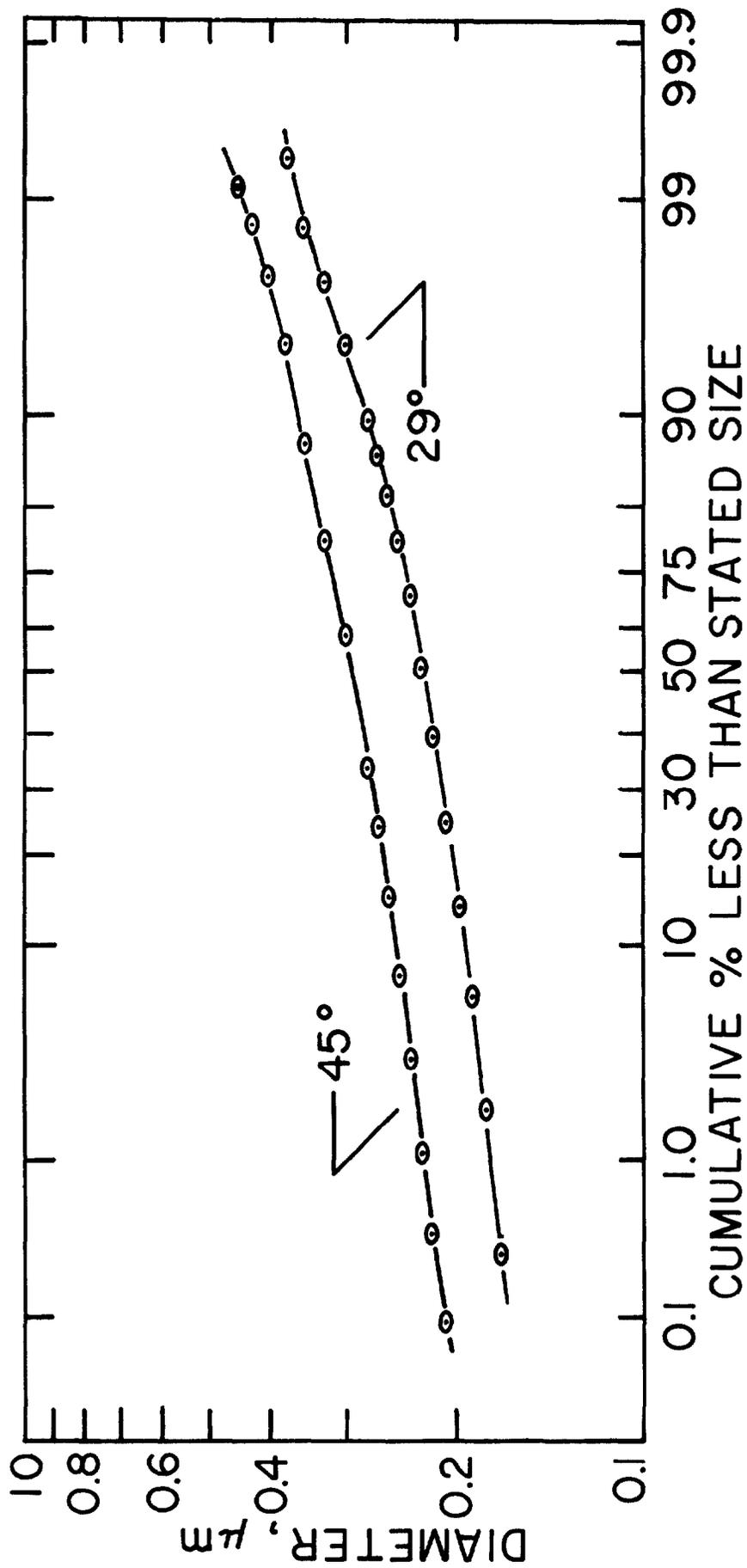


Fig. 5. Cumulative size distributions of DOP aerosol produced by Q-127 generator at analyzer settings of 29° and 45°.

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Table 4. Refractive index of PSL and DOP from four sources at 589 nm.

<u>Source</u>	<u>Refractive Index at 22°c</u>
PSL	1.5905
"New DOP " BASF	1.4850
"New DOP " Kodak	1.4845
"Old DOP"	1.4858
"Aerosol DOP"	1.4864

Discussion

Table 5 shows filter penetration results for identical filter papers tested at 5 different laboratories.⁵ All measurements were made with the mechanical analyzer set to 29° except for the measurements shown in the first column for which the analyzer was set to 45°. There is significant variability in results between laboratories except that Dexter and Hollingsworth and Vose show close agreement. The penetration results obtained at Harvard when using an analyzer setting of 45° are consistently less than the results at 29°. The average ratio of penetration at 29° to 45° is 2.50 with a range from 1.84 to 3.25. For both analyzer settings, the test aerosols have the same geometric standard deviation but the median size differs by 0.07 µm. Thus a 23% reduction in median particle diameter produces a 250% increase in penetration for the DOP size range between 0.23 and 0.3 µm.

The decrease in penetration with increasing particle size suggests that impaction is the primary removal mechanism. If the test aerosol particles were to be reduced to a size close to that for maximum penetration, the effect on penetration of small size variations would be minimized.

With one or two exceptions, the two Harvard readings for analyzer settings of 29° and 45° bracket the penetration measurements made at other laboratories. This suggests that if the test aerosols used at these other laboratories had the same GSD as Harvard's (i.e., 1.15) the differences in CMD between laboratories would have to cover a range as large as 30%. As will be discussed later, these differences in CMD at constant GSD can be produced only at analyzer settings significantly different than 29°. Inasmuch as the standard operating condition for the Q-127 is an analyzer setting of 29°, the observed differences in filter penetration must be a result of differing geometric standard deviations of the test aerosols.

The Harvard data points in Figure 6 show approximately the same shape as the theoretical line calculated by Sinclair³ for a refractive index of 1.483 except that the particle diameters are 13 to 29% less at the same analyzer angle. It must be kept in mind, however, that the Harvard data points represent count median diameters of polydisperse aerosols whereas Sinclair's theoretical polarization ratio line was determined for perfectly monodisperse aerosols. The aerosols produced by the Q-127 cover a significant range of sizes, e.g., for a GSD of 1.15, having a two standard deviation range of +35% and -24% of the CMD. Therefore, the mechanical analyzer is measuring an

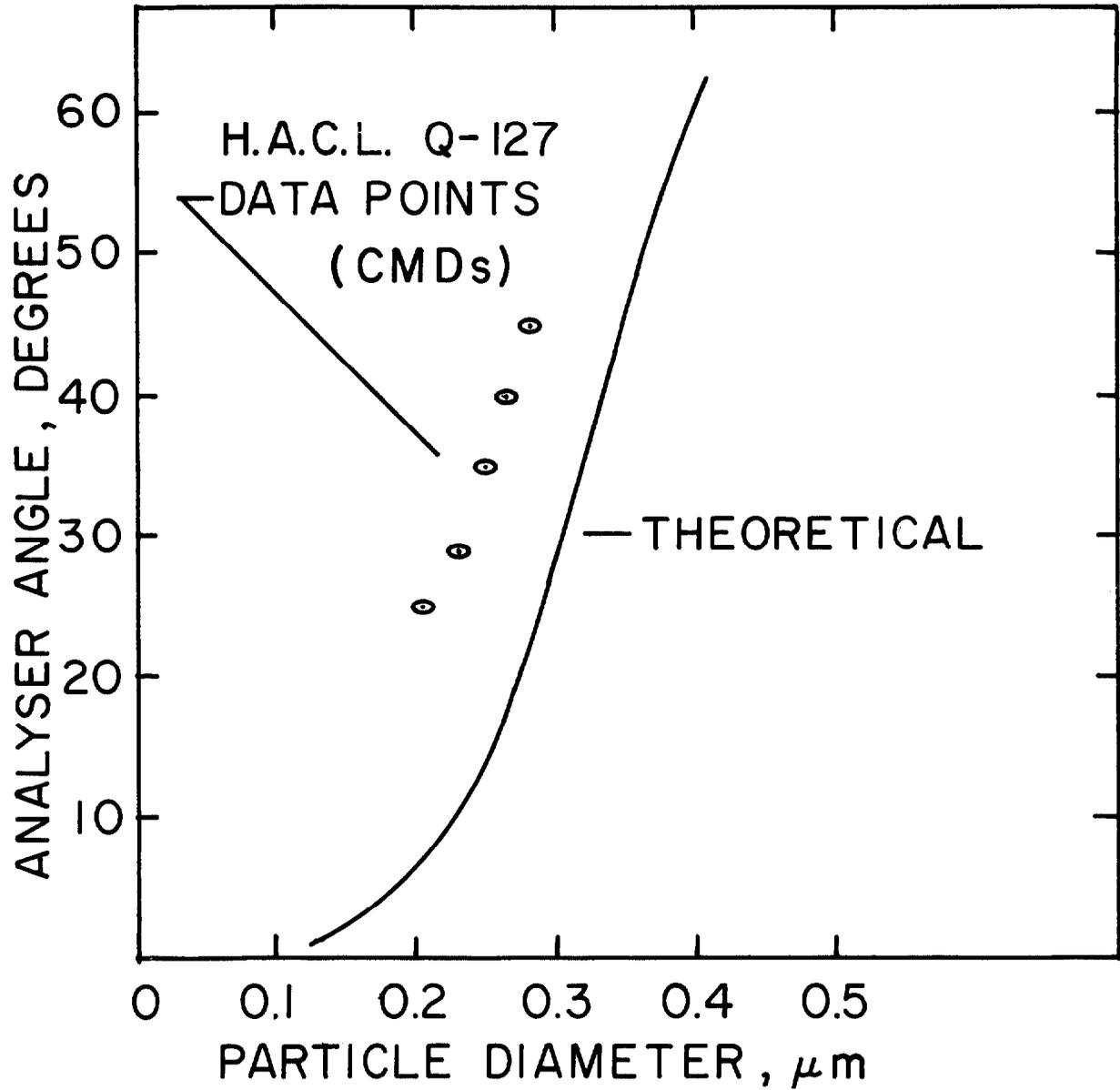


Fig. 6. Comparison of theoretical and measured particle size versus polarization ratio. (Theoretical curve from Sinclair,³ interpolated for $m = 1.483$.)

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average polarization ratio for these polydisperse aerosols. To determine the significance of polydispersity for this measurement, a graph of polarization ratio, i_2/i_1 , versus particle size was constructed in Figure 7 by converting analyzer angle, ϕ , as given by Sinclair³ and shown in Figure 6, to polarization ratio by the relationship,

$$i_2/i_1 = \tan^2\phi. \quad (1)$$

Over the size range 0.15-0.4 μm , polarization ratio can be approximated by a constant power function of diameter,

$$i_2/i_1 = 4730 d^{8.1}, \quad (2)$$

for diameter in μm .

Table 5. DOP penetration measured at five laboratories.⁵

Sample	Harvard (45°)	Harvard (29°)	Dexter (29°)	H&V (29°)	Fiber Process (29°)	Rocky Flats (29°)
18	.018	.041	.028	-	-	.022
30	.016	.038	.028	-	-	.0215
14	.010	.027	.021	-	-	.015
11	.004	.012	.009	-	-	.006
23	.001	.002	.002	-	-	.001
25	.012	.022	.019	-	-	.014
34	.004	.012	.009	-	-	.008
12	.007	.018	.013	-	-	.009
S 20	.035	.068	.046	.049	.053	-
S 21	.007	.016	.013	.014	.015	-
S 22	.025	.052	.038	.039	.042	-
S 23	.005	.010	.007	.008	.009	-
S 24	.006	.014	.013	.014	.019	-
1A	.008	.018	.013	.015	-	-
6A	.004	.013	.009	.010	-	-
20A	.010	.032	.028	.027	-	-
21A	.005	.016	.011	.012	-	-
23A	.003	.009	.008	.008	-	-
24A	.010	.024	.017	.017	-	-
25A	.012	.029	.021	.021	-	-
28A	.006	.016	.011	.012	-	-
29A	.002	.005	.005	.004	-	-
30A	.016	.035	.025	.026	-	-

Analyzer setting shown in parenthesis.

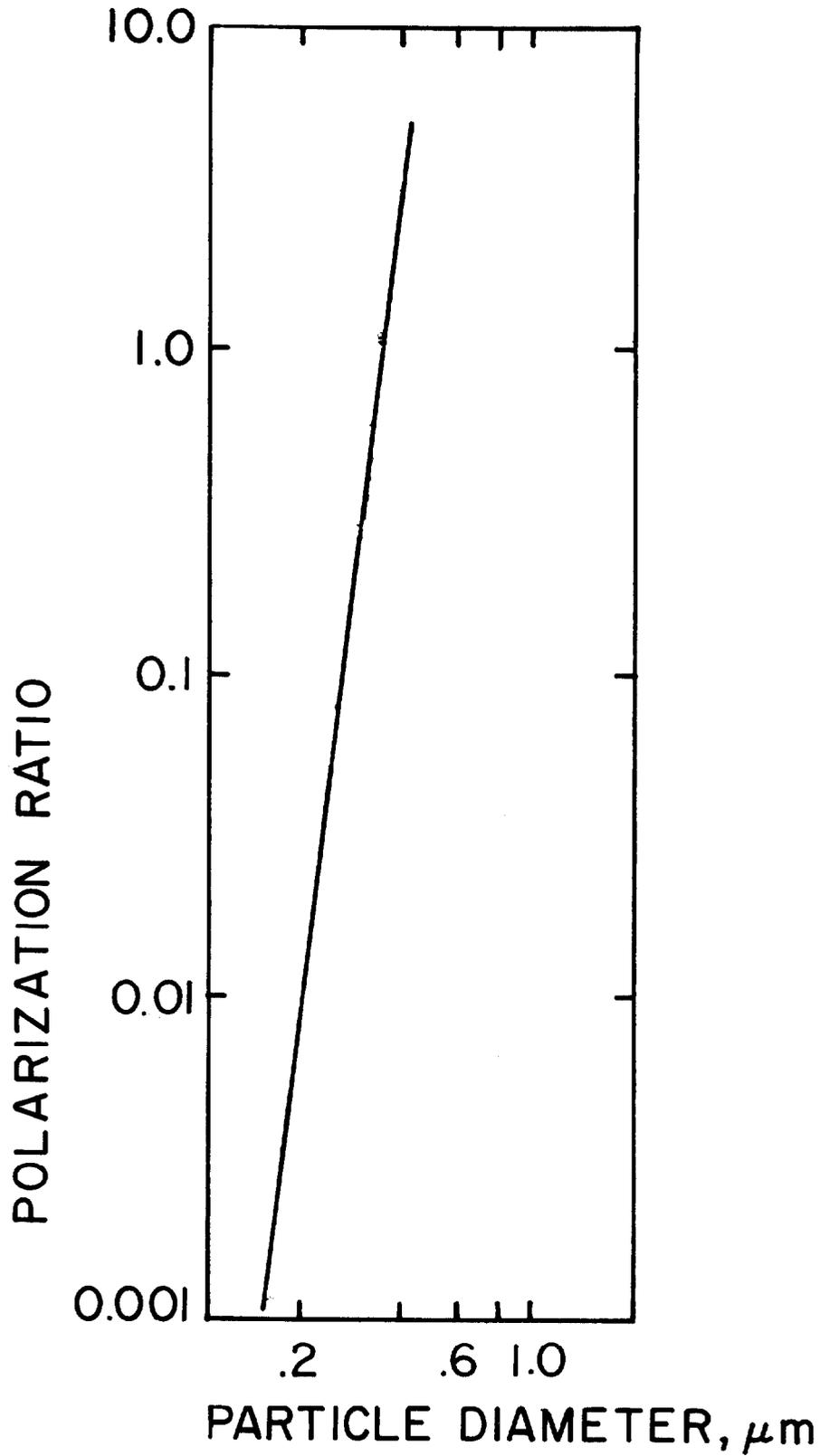


Fig. 7. Polarization ratio versus particle diameter - calculated from Sinclair's curve of analyzer angle vs. diameter³ for $m = 1.483$.

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The mechanical analyzer measures the average polarization ratio which is interpreted in practice as representing the diameter of aerosol particles of uniform size using Sinclair's³ curves of analyzer angle versus monodisperse particle size.

The average polarization ratio is defined as the ratio of the intensities of the horizontal and vertical polarization components of 90° scattered light. For a polydisperse aerosol the average polarization ratio is given by,

$$\overline{PR} = \frac{I_2}{I_1} = \frac{\sum n_j(i_2)_j}{\sum n_j(i_1)_j} \quad (3)$$

and the diameter of average polarization ratio by,

$$d_{\overline{PR}} \approx \left[\frac{\sum n_j(i_2)_j}{\sum n_j(i_1)_j} \right]^{\frac{1}{8.1}} \quad (4)$$

where n_j is the number of particles in the j th size category, i_2 and i_1 are the intensity parameters for horizontal and vertical polarization, respectively, and the summations are for all particle size categories. Values for i_1 and i_2 were calculated with a computer program⁽⁸⁾ and integrated over scattering angles from 88.5° to 91.5° and wavelengths defined by the emission characteristics of the light source (tungsten filament), the transmission of a number 59 wratten filter, and the sensitivity of the S-4 photodetector. Results of calculations using equation (3) are shown in Table 6 for hypothetical aerosols all of which may be produced at an analyzer setting of 29° and interpreted as $d_{\overline{PR}} = 0.3 \mu\text{m}$. Table 6 shows that at a GSD of 1.05, the condition for which Sinclair originally used the OWL ($0.3 \mu\text{m} \pm 10\%$ is taken to mean ± 2 standard deviation), the CMD and $d_{\overline{PR}}$ are indistinguishable.

Table 6. CMD of aerosols with log normal size distribution produced by HOT DOP generator at a mechanical analyzer setting of 29°.

<u>GSD</u>	<u>CMD</u>
1.00	0.300
1.05	0.294
1.10	0.286
1.15	0.274
1.20	0.260
1.25	0.246
1.30	0.230

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The size distributions measured at Harvard with the PMS analyzer had GSDs of about 1.15. Reference to Table 6 indicates that the GSD does not fully account for the entire difference between the average polarization size of 0.3 μm and the measured size of 0.23 μm . However, an examination of the size distribution for analyzer angles of 29° and 45°, Figures 4 and 5, reveals that the distributions are not perfectly log normal but were slightly enriched at the large particle end of the distribution. Consequently, a more accurate estimate of the effect of polydispersity was made by numerical integration of equation (4) for the actual size distribution obtained at analyzer settings of 25°, 29°, 35°, 40°, and 45°. Results, shown in Table 7, show good agreement between particle size obtained from Sinclair's curve (Figure 6) and the diameter of average polarization ratio calculated from actual size distributions by numerical integration.

Table 7. Results of numerical integration of measured size distributions to obtain diameter of average polarization ratio.

Analyzer Setting	Measured CMD	Calculated Dia. of Average Polarization Ratio	Dia. from Sinclair's Curve, Figure 6
25°	0.205	.270	.290
29°	0.232	.278	.302
35°	0.250	.290	.318
40°	0.265	.302	.334
45°	0.300	.323	.346

One can conclude from this that the differences between 0.3 μm and the measured values of CMD are due primarily to the effects of polydispersity, which is significant, even at these narrow distributions, because the light scattering weighted diameter of average polarization ratio is such an extremely strong function of particle size (i.e., $d^{0.1}$).

Conclusions

- 1) The GSD of DOP aerosol produced by a Q-127 generator has been measured to be about 1.15. The GSD is sufficiently large to affect the interpretation of data from tests designed to use monodisperse aerosol.
- 2) The fact that the CMD is less than 0.3 μm when the analyzer setting is 29° is the result of aerosol polydispersity.
- 3) Filter penetration is sensitive to particle size because filter operating conditions are in the impaction region. It was thought when 0.3 μm was selected for the test aerosol for HEPA filters that this diameter was the size for maximum penetration and that the sensitivity of penetration measurements would be little affected by small size changes. Later studies showed the size for minimum penetration to be 0.08 μm and the size-penetration curve to be very steep in the region of 0.3 μm .

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- 4) Therefore, when a DOP test aerosol is produced at an analyzer setting of 29° , small variations in GSD produce significant variations in filter penetration unless GSD is less than 1.09. Inasmuch as measured GSD's are closer to 1.15, variations in GSD are believed to be the cause of interlab variability in penetration measurements.
- 5) The following steps would reduce interlab variability:
 - a) Use a test aerosol of smaller particle size that corresponds more closely to the size for maximum penetration.
 - b) Continue to use the 29° polarization ratio but restrict GSD to <1.09 . This corresponds to a CMD of 0.285, within 5% of $0.30 \mu\text{m}$ but means that a method must be devised to produce such an aerosol and to verify that the GSD is <1.09 .
 - c) Continue to use the 29° standard but specify that the GSD must be within $\pm .01$ of 1.15. This also means that a method must be devised to monitor GSD accurately at each laboratory.
 - d) Continue to use the standard 29° setting on DOP generators and apply a correction factor to penetration readings that are based on a comparison of filter readings with a single reference lab that all testing laboratories employ for this purpose.

Acknowledgement

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Appendix C FIBER DIAMETER DETERMINATION OF FIBER GLASS SAMPLES

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Abstract

The Kozeny-Carmen equation is the basis of present practice in relating fiber diameter to handsheet ΔP in quality control of fiber glass papers used for filtration. The Naval Research Laboratory developed the following equation relating volume-surface diameter to the measured ΔP :

$$D = \frac{2.3}{\left\{\frac{\Delta P}{L}\right\}^{1/2}} \quad \begin{array}{l} \Delta P = \text{mm H}_2\text{O} \\ L = \text{inches sheet thickness} \\ D = \text{Volume-surface diameter in microns} \end{array}$$

This equation assumes a constant linear velocity, a constant porosity in the glass sheet and a constant shape factor in the Kozeny-Carmen equation. We have found that porosity and shape factor vary considerably depending upon preparation techniques and fiber diameter distribution. Thus, direct correlations of handsheet ΔP to fiber diameter can result in considerable error by present practice.

The BET surface area provides us with the most accurate and reproducible and probably the most useful technique for fiber diameter determination. Using this method of surface area measurement, we have found good correlation between handsheet ΔP and surface area only if the effects of porosity are taken into consideration.

By using the Frazier air permeability tester, the volume surface fiber diameter (BET) is correlated to air flow and porosity using the following equation:

$$D = C \left\{ \frac{Q^2 W^3}{L} \right\}^{1/4}$$

where:

- Q = flowrate of air (STP) $\text{ft}^3/\text{ft}^2/\text{min}$
- W = handsheet sample weight in grams
- L = handsheet caliper in mils
- C = a constant for this test (0.176)

Introduction

The single most important parameter of glass microfiber products is the average fiber diameter of a given lot. The various codes are designated by ranges of fiber diameter and are priced accordingly. There are several different definitions for "average" fiber diameter, but the one of most consequence to our customers is the volume-surface average diameter. This is defined as the diameter that a theoretical fiber would require so that it would have the same surface area per unit weight as an actual fiber sample. Consequently, all of our reference fiber diameters were derived from

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surface areas obtained using BET krypton adsorption.

Currently, the Williams Freeness Test is used to run regular Quality Control checks on microfiber production. This method empirically relates the drainage time of a fiber slurry of known pH, temperature and concentration to its volume-surface average diameter. Several recent refinements on this test procedure have perfected it to a point where results on any given sample of fiber can be reproduced within ± 1.0 percent.

In spite of the precision in this method, it has become strikingly obvious that two different samples can have identical Williams Freeness drain times yet have different volume-surface average fiber diameters. This point indicates that liquid drainage, in the form of the Williams Freeness Test, is dependent on more than just the volume-surface fiber diameter. We believe that some of the error involved in drainage tests could either be accounted for or eliminated in a refined ΔP test, using air permeability as an indirect measurement of surface area. This concept is already in use in various test equipment, such as the Fisher Sub-Sieve Sizer, the Blaine Permeability Tester, and the Scheffield Micronaire Tester. The ΔP test is also used as a Quality Control Test for air filtration blankets. The intention of the paper is to refine the basic principle of these ΔP tests into a form that will give a true indication of the volume-surface average fiber diameter.

Discussion

The basic law governing one-dimensional flow through a permeable media is given by Darcy to be

$$\frac{Q}{A} = \frac{B_0 \Delta P}{u L}$$

Q = flowrate
A = area of sample
B₀ = permeability coefficient
u = fluid viscosity
L = media thickness

This equation is applicable in cases of viscous streamline and laminar flow of liquids, and applies to gases when ΔP is small when compared to P, the ambient pressure. B₀, however, is not a constant, and is generally expressed as a function of E, the porosity, S₀, the surface area per unit volume of the sample, and k, an empirical constant. There have been numerous solutions to this function but we shall deal with only two.

The most famous treatment of this equation is the Kozeny-Carman form. This is a theoretical formulation based on a relationship analogous to the Poiseuille law of flow through a circular pipe, except that the hydraulic radius is used in the equation. The hydraulic radius is defined as the volume of sample filled with liquid divided by the wetted surface. Using this principle, the Kozeny-Carman equation takes the form:

$$B_0 = \frac{1}{K S_0^2} \frac{E^3}{(1-E)^2}$$

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$$\text{and } \frac{Q}{A} = \frac{\Delta P}{K S_o^2 uL} \frac{E^3}{(1-E)^2}$$

For non-porous fiber samples, $S_o = 4/D$, where D equals the volume-surface average fiber diameter. Therefore, the Kozeny-Carman equation can be re-written in the form

$$D = \left[\frac{16 Q k u L (1-E)^2}{A \Delta P E^3} \right]^{1/2}$$

where D is determined solely in measurable quantities.

There are certain limitations to this equation, however. First, the equation was derived for randomly packed samples. Fibers in most samples tend to be oriented, often severely so. Also, this treatment assumes a uniform pore size, whereas there is actually a distribution of pore sizes. But of most importance is the relationship of k to the porosity, E. For oriented fiber beds, k remains constant only when E is less than 0.8. As E approaches 1.0, k rises rapidly. Table I gives some typical values of E versus k.

The Naval Research Laboratory developed a modification of this approach. A handsheet of the fiber in question is subjected to air with a constant linear velocity Q/A (14.2 cm/sec) and the resulting pressure drop is measured. By assuming a constant E for identically prepared samples, NRL reduced the Kozeny-Carman equation to:

$$D = \frac{2.3}{\frac{\{\Delta P\}}{L}}^{1/2} \quad \begin{array}{l} \{P\} = \text{mm H}_2\text{O} \\ \{L\} = \text{mils} \\ \{D\} = \text{microns} \end{array}$$

This equation in effect assumes that the combination of terms $k(1-E)^2(E)^{-3}$ is constant. Looking at the third column in Table I, we see that this is not the case. This indicates that the NRL equation is valid only when the porosity of the sample is the same as NRL assumed value. However, even identically prepared samples can have varied porosities.

There is also an empirical solution to the Darcy permeability coefficient. Davies has determined that for beds of fibers with porosities between 0.7 to 0.99:

$$B_o = \frac{D^2}{64 (1-E)^{1.5} \{1 + 56 (1-E)^3\}}$$

Furthermore, for E greater than 0.9, B_o is closely approximated by:

$$B_o = \frac{D^2}{64 (1-E)^{1.5}}$$

This equation does not contain K. When combined with the Darcy equation we obtain:

$$D = \left\{ \frac{64 Q u L (1-E)^{1.5}}{A \Delta P} \right\}^{1/2}$$

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Since this is an empirical solution, its accuracy depends on the method of obtaining the reference diameters. We found that a better correlation with BET - determined fiber diameters is obtained by replacing the "64" with "36." The diameter is then given by:

$$D = \left\{ \frac{36 Q u L (1-E)^{1.5}}{A \Delta P} \right\}^{1/2}$$

This equation allows calculation of the fiber diameter from strictly measurable quantities by fixing either Q or ΔP and measuring the other. The Naval Research Laboratory chose to fix the flowrate and measure ΔP . However, we chose to fix the pressure differential and measure the flowrate, using a Frazier Permeability Tester adapted with a 1 square inch, wire mesh-supported opening. There are several reasons for this decision:

1. The calculated diameters are very sensitive to error in pressure readings at low values of ΔP (less than 50 mm H₂O). This sensitivity decreases as ΔP increases. Using the Frazier Tester, a ΔP can be chosen that is above this sensitive region. For this work we chose a ΔP equal to 5 inches (127 mm) H₂O, which is roughly in the middle of the range of ΔP for the NRL test. The wide range of flowrates encountered can be measured readily on the Frazier Tester by changing the calibrated orifices in the tester.
2. The Darcy equation for flow through porous media needs a correction factor for compressible gases,

$$\text{Correction Factor} = \frac{P_1 + 1/2 \Delta P}{P_1}$$

(the average pressure through the medium divided by the ambient pressure), which is usually assumed to be negligible for small ΔP . By fixing the ΔP at a specified value, this term becomes a constant and the effects of ignoring it in the theory can be accounted for in an equipment calibration constant.

3. The wide range of measurement possible using the different orifices allows considerable latitude in specifying other test parameters. The most notable of these is that thicker samples were used (approximately four times that of a standard handsheet) to minimize the effects of error in measuring L , the sample caliper.

The test samples were prepared by running a revised William Freeness Test on each sample of fiber. After measuring the Williams Freeness time, the samples were allowed to finish draining, and then were removed from the screen using blotter paper, pressed with a roller similar to a British Handsheet roller, and then allowed to dry. The actual test samples were 2.25 inch diameter disks that were punched from the center of the Williams Freeness pads to minimize the effect of fiber build-up around the outside of the Williams Freeness Tester. The samples were weighed, measured for caliper on a bigfoot micrometer with light downward pressure to minimize compression, and then measured on the Frazier tester. The data was analyzed using several methods that are discussed in the next section. An outline of our

procedure is included in Appendix I.

Results

The primary difference between the Kozeny-Carman (NRL modification) and the Davies solutions is how they deal with the effects of porosity. The NRL solution assumes all fiber samples would have the same porosity if prepared in identical manner, regardless of the average diameter. However, the packing of fiber is greatly affected by the fiber diameter distribution. Samples with broad distributions should pack more tightly than those with narrow distributions. Therefore, if the resulting change in porosity is not accounted for, the results of a ΔP fiber diameter determination would be in error.

It was necessary to test whether the fiber diameter distribution has a real and significant effect on ΔP measurements. The actual fiber diameter distribution is difficult to measure, so we prepared a series of samples with progressively broader relative distributions. Samples of Codes 100, 106 and 110 fiber were chosen so that a blend of Codes 100 and 110 would have the same volume-surface average fiber diameter as the Code 106. Samples of 106 with increasing proportions of 100/110 mixture were prepared so that they would have the same volume surface fiber diameter with progressively broader distributions. Table II shows the results of these tests using both equations to evaluate the data.

BET determined fiber diameters confirmed that the blends used in this series did in fact have the same average diameters. The effect of fiber diameter distribution on the porosity, E , is shown. As expected, E decreases as the distribution broadens. This is reflected in the Williams Freeness data. Our samples had diameters of 0.70 microns, but the Williams Freeness diameters for the artificially broadened samples dropped as low as 0.65 microns. Also, it must be noted that our sample of 100% Code 106 had a diameter distribution of its own, so that it is quite possible that a 0.70 average diameter fiber sample with a narrower distribution could appear to be even coarser than 0.70 microns on a Williams Freeness Tester. Therefore, unless there is additional information about fiber diameter distribution or porosity, the Williams Freeness Test is only valid as an approximation.

The same samples were then checked using the conditions specified by the NRL method. The resulting fiber diameters are also listed in Table II. They show the same trend as the Williams Freeness data; narrow distributions cause the NRL ΔP test to indicate larger diameters than do broad distributions, even though the samples have the same volume-surface average diameter. Therefore the NRL test is susceptible to the same variation in sample diameter distribution as the Williams Freeness Test.

The last column shows the effect of accounting for the change in porosity caused by varying diameter distributions. This data, analyzed using the Davies equation, does not exhibit the same distinct relationship between diameter and distribution as the first two methods. (Figure I). The scatter among the data points appears to be more pronounced for the Davies equation, probably due to the

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additional measured parameters. However, the standard deviation was 2.8 percent for the Williams Freeness data, 6.3 percent for the NRL ΔP data, and 1.5 percent for the data analyzed with the Davies equation. Clearly, the possible increase in experimental error using the Davies equation is more than offset by reducing the error caused by the assumption of constant porosity. It should be noted that these results are consistent with results obtained on handsheets made on a British mold and a Woods and Noble mold, but that use of Williams Freeness disks produced more consistent data.

To determine how well the Kozeny-Carman and the Davies equation correlate with BET results over a wide range of fiber diameters, uniform samples of Codes 100, 102, 104, 106, 108A, 108B, and 110 were prepared by dispersing small bits of each code in a turbulent airstream. Random samples of each were measured on BET equipment to determine their volume-surface fiber diameters and to confirm their homogeneity. Triplicate samples of each grade were prepared on the Williams Freeness Tester and then tested on the Frazier Permeability Tester. The data was analyzed with the Davies equation and with the full Kozeny-Carman equation. Table III shows the mean fiber diameter of each code determined by these methods and their corresponding surface areas. ($d = 1.53/S.A.$). There does not appear to be a good correlation of either the Kozeny-Carman or the Davies diameters with the BET diameters. The reason for this can be seen by plotting surface areas for each against the BET surface areas. Both curves are linear to $1.5 \text{ m}^2/\text{g}$, which corresponds to about 1 micron. However, both fall off as the fiber samples become finer, suggesting that neither method of analyzing allows for direct calculation of surface areas and corresponding fiber diameter less than one micron. It is necessary to use a graphical correlation between air permeability diameters and BET diameters similar to the one relating Williams Freeness times to BET diameters. While either the Davies or the Kozeny-Carman equation could be used for this purpose, the simplified form (Appendix II) of the Davies equation was chosen because it is much easier to work with.

$$D = 0.176 \left\{ \frac{Q^2 W^3}{L} \right\}^{1/4}$$

D = fiber diameter, microns
Q = flowrate, $\text{ft}^3/\text{ft}^2\text{-min}$
W = sample weight, grams
L = caliper, inches

The bracketed term $\left\{ \frac{Q^2 W^3}{L} \right\}$ has a one-to-one correspondence to the fiber diameter and is called the Frazier number. These numbers were calculated for the seven fiber Codes and are listed in Table IV. By plotting Frazier numbers against BET fiber diameters, a calibration curve is obtained (Figure III). Therefore, by measuring the weight and caliper of, and flowrate through, a prepared fiber sample, and calculating the corresponding Frazier number, the volume-surface average fiber diameter can be obtained graphically without error caused by differences in porosity and fiber diameter distribution.

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Conclusions

From this work we can conclude the following:

1. The porosity of a bed of fibers must be accounted for in determining the volume-surface average fiber diameter using fluid flow techniques (drainage or air permeability).
2. Neither the Williams Freeness Test nor the Naval Research Laboratories ΔP test adequately accounts for the variation in fiber diameter distribution inherent in production fiber samples.
3. Evaluations using the Davies modification of the Darcy equation does account for the variation in fiber diameter distribution.
4. The primary term in the Davies equation, $\frac{Q^2 W^3}{L}$, can be correlated graphically with the BET derived volume-surface average fiber diameters.

Appendix I

Scope

This method is used to determine the true volume-surface average fiber diameter unbiased by diameter distribution effects. This value can be compared to the Williams Freeness value obtained during the procedure.

Equipment

1. Williams Freeness Tester with Timer and 80 mesh screen plate (Williams Apparatus Co., Watertown, N.Y.).
2. Balance, sensitive to ± 0.01 g.
3. Waring Blender, Model 1120.
4. Thermometer.
5. Deionized water.
6. 10 ml graduate.
7. 0.1N NaOH.
8. Blotter paper, 4 in. x 4 in.
9. British Handsheet Roller or Equivalent.
10. 2.25 in. I.D. circular punch.
11. Micrometer, sensitive to 0.5 mils.

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12. Frazier High Pressure Differential Model Air Permeability Tester with 1 in.² adapter plates with open mesh screen support. (Frazier Precision Instrument Co., Gaithersburg, Maryland).
13. Small circle of 8 mesh screening to fit the bottom of the Williams Freeness Tester.

Procedure

1. Weigh 1.00 ±0.01 gram of clean, dry fiber. For fiber diameters greater than 1.01 microns, weight out 2.00 ±0.01 grams of fiber.
2. Pull the weighed fiber apart into several pieces and place in a clean empty Waring Blender jar.
3. Add approximately 500 ml. deionized H₂O to the jar, and then add 10 ml. of the 0.1N NaOH.
4. Turn blender on high speed and beat the fiber slurry for exactly two minutes.
5. Place the small screen in the cone of the tester. With the valve closed, fill the cone with deionized water until a meniscus forms above the base. Drop the screen plate onto the meniscus, taking care that no bubbles form beneath the screen, then close the tester and clamp in place.
6. Add approximately 100 ml. H₂O to the tester to cover the screen. Then transfer the fiber slurry to the tester, using small amounts of deionized H₂O to rinse all fiber from the Waring blender lid and jar. Add additional H₂O to the tester to bring the slurry level to the 1000 ml. mark.
7. Stir the slurry slightly to assure good dispersion of the fiber. Check slurry temperature (should be 68-72°F).
8. Reset timer to ZERO.
9. Open the drain valve rapidly. When slurry level reaches the zero mark, close the drain valve rapidly.
10. Record elapsed time noted on the timer. (This time can be converted to fiber diameter by use of calibration graphs).
11. Reopen valve to allow fiber slurry to finish draining. Open the tester and remove excess H₂O from the pad using blotting paper and hand pressure.
12. Separate the pad from the screen and place between fresh sheets of blotter paper. Press with the roller using three passes, then dry the samples (with blotter paper) in an oven. The fiber pads will separate from the paper as they dry.
13. Remove a disk from the center of the fiber pad using the 2.25 in. circular punch.

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14. Weigh the disk to the nearest 0.01 gram and measure its caliper to the nearest 0.5 mil.
15. Place disk over the wire mesh-backed 1 square inch opening in the Frazier Tester. Clamp tightly.
16. Install an orifice in the tester. The appropriate size orifice is determined by trial and error for each grade of fiber.
17. Start the motor and increase its speed by means of the Variac control until a reading of 5.0 inches H₂O is obtained on the left hand manometer, while making sure that the right hand manometer does not overflow. The right hand manometer should read between 4 and 20 inches of manometer oil (Sp. Gr. =0.827) for most accurate results. If not, a different orifice should be selected.
18. Once the right hand manometer reading is obtained, its corresponding flow rate is obtained from the orifice calibration tables.
19. Calculate the following value.

$$N = \frac{\left\{ \frac{\text{ft}^3}{\text{min-ft}^2} \right\}^2 \text{ weight (g)}^3}{\text{Caliper (inches)}}^3$$

20. Refer to Figure III to determine the average fiber diameter corresponding to the calculated value, N.

Notes:

1. A separate calibration curve should be prepared relating BET diameter to ΔP results for each specific set of test equipment.
2. All openings on the Frazier Tester should be sealed, the orifices should be tight, the tester should be level and the manometers should be filled to zero points before running samples.

Appendix II Derivation of final forms of Kozeny-Carman and Davies equations.

The difference between the Kozeny-Carman equation and the Davies equation lies in their expression of B_o, the permeability coefficient in the Darcy equation.

1. Kozeny-Carman $B_o = \frac{d^2}{16K} \frac{E^3}{(1-E)^2}$
2. Davies $B_o = \frac{d^2}{36 (1-E)^{1.5} \{1+56 (1-E)^3\}}$

As E, the porosity, becomes large, above 0.9, the Davies expression is closely approximated by:

3. $B_o = \frac{d^2}{36 (1-E)^{1.5}}$

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The porosity is defined as the ratio of the volume of free space in a sample to the total volume of the sample. This can be expressed as:

$$4. \quad E = \frac{AL - W/\rho}{AL}$$

A = area of sample
 L = thickness of sample
 W = weight of sample
 ρ = density of sample material

Substituting equation 4 into 1 and 3 we obtain:

$$\text{Kozeny-Carman} \quad B_o = \frac{d^2 (AL - W/\rho)^3}{16 K A L (W/\rho)^2}$$

$$\text{Davies} \quad B_o = \frac{d^2}{36} \left(\frac{AL}{W/\rho} \right)^{1.5}$$

When substituted into the Darcy equation and solved for the diameter, we obtain:

$$\text{Kozeny-Carman} \quad d = \left\{ \frac{16 Q K A L^2 \eta (W/\rho)^2}{A^1 \Delta P (AL - W/\rho)^3} \right\}^{1/2}$$

$$\text{Davies} \quad d = \left\{ \frac{36 Q \eta L}{A^1 \Delta P} \frac{W/\rho}{AL} \right\}^{1.5} \quad 1/2$$

where d = diameter of fiber, microns
 Q = volumetric flowrate, ft³/ft² min
 K = constant (Kozeny-Carman only)
 A = area of sample, 25.65 cm²
 L = caliper of sample, inches
 W = weight of sample, grams
 ρ = density of sample material, g/cc
 η = viscosity of air at room temperature, 1.84×10^{-4} poise
 A^1 = area of sample tested for air permeability, 1-in.²
 ΔP = pressure drop across the sample, 5-in. H₂O

Using the numerical values specified in the above list and appropriate conversion factors, these equations reduce to:

$$\text{Kozeny-Carman} \quad d = 9.05 \left\{ \frac{Q W^2 L^2}{6.45 L - (W/\rho)} \right\}^{1/2}$$

$$\text{Davies} \quad d = 0.362 \left\{ \frac{Q^2 W^3}{L \rho^3} \right\}^{1/4}$$

For 475 glass, ρ equals 2.61 g/cc. Therefore, when applied to our glass microfibers, the Davies equation can be written:

$$d = 0.176 \left\{ \frac{Q^2 W^3}{L} \right\}^{1/4}$$

Notes:

1. The background for these equations can be found in The Flow of Gases Through Porous Media; Carman, P.C., Academic Press, Inc.

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N.Y., 1956.

- The value of K used in the Kozeny-Carman equation was 4.25, designated as the "shape factor" in the NRL adaption.

Appendix III Table I Typical k versus e for porous media

\bar{E}	\bar{K}	$\frac{K(1-E)^2}{E^{-3}}$
0.6	4.3	3.18
0.7	4.4	1.15
0.8	5.4	0.422
0.9	8.8	0.121
0.95	15.2	0.044
0.98	32.8	0.014
0.99	61.0	0.006

Table II

Composition			Porosity	Willaims Freeness	NRL ΔP	Davies
% 100	% 106	% 110	E	Diameter	Diameter	Diameter
0	100	0	0.928	0.705microns	0.805microns	0.780 mic.
9	80	111	0.924	0.690	0.771	0.772
18	60	22	0.919	0.680	0.739	0.802
27	40	33	0.916	0.665	0.727	0.795
36	20	44	0.911	0.660	0.684	0.802
45	0	55	0.908	0.650	0.673	0.780

Table III

Code	BET		Kozeny-Carman		Davies	
	Diameter	Surface Area	Diameter	Surface Area	Diameter	Surface Area
100	0.343	4.46 m ² /g	0.41mic.	3.71 m ² /g	0.50mic.	3.06 m ² /g
102	0.401	3.81	0.43	3.59	0.54	2.83
104	0.520	2.94	0.52	2.94	0.65	2.35
106	0.697	2.19	0.62	2.54	0.78	1.96
108A	0.992	1.54	0.76	2.00	1.00	1.53
108B	1.86	0.82	1.36	1.12	1.83	0.84
110	3.93	0.39	2.74	0.56	3.58	0.43

Table IV

Code	BET Fiber Diameter	Frazier Number
100	0.34 microns	65.6
102	0.40	88.0
104	0.52	186
106	0.70	380
108A	0.99	1050
108B	1.86	11,600
110	3.87	169,000

Figure I Distribution versus fiber diameter davies, NRL ΔP , Williams Freeness

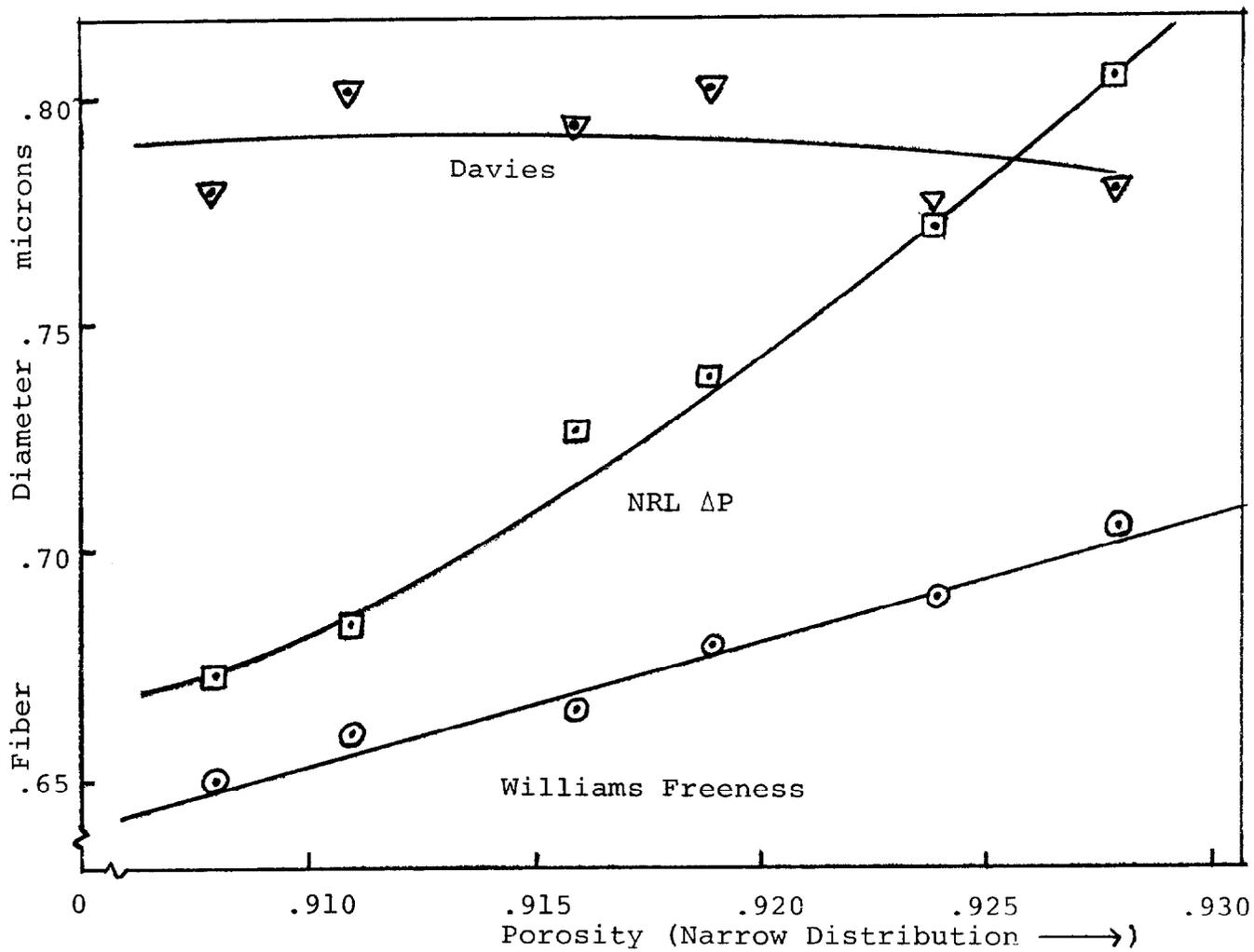


Figure II Surface Area, BET versus air permeability

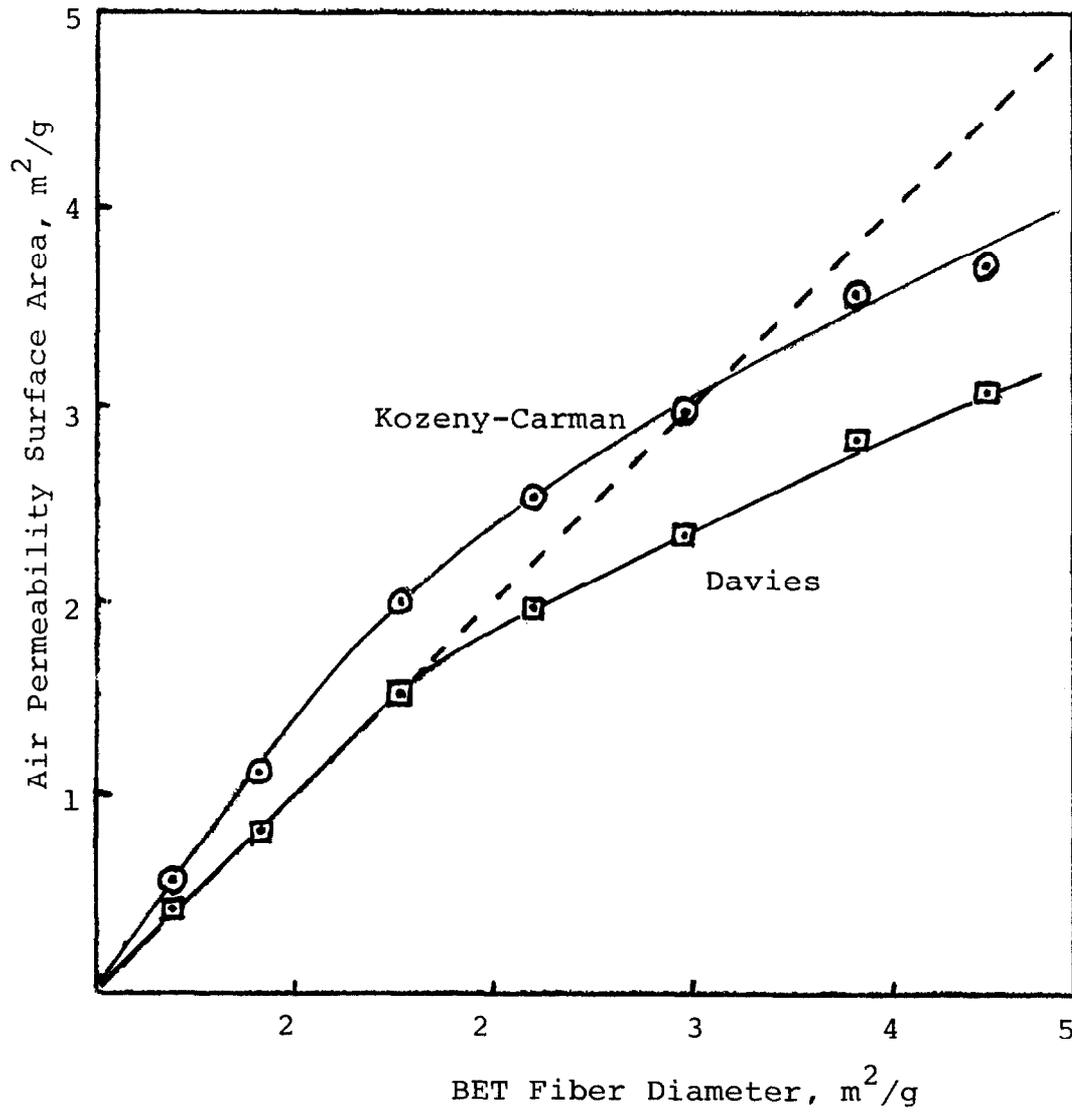
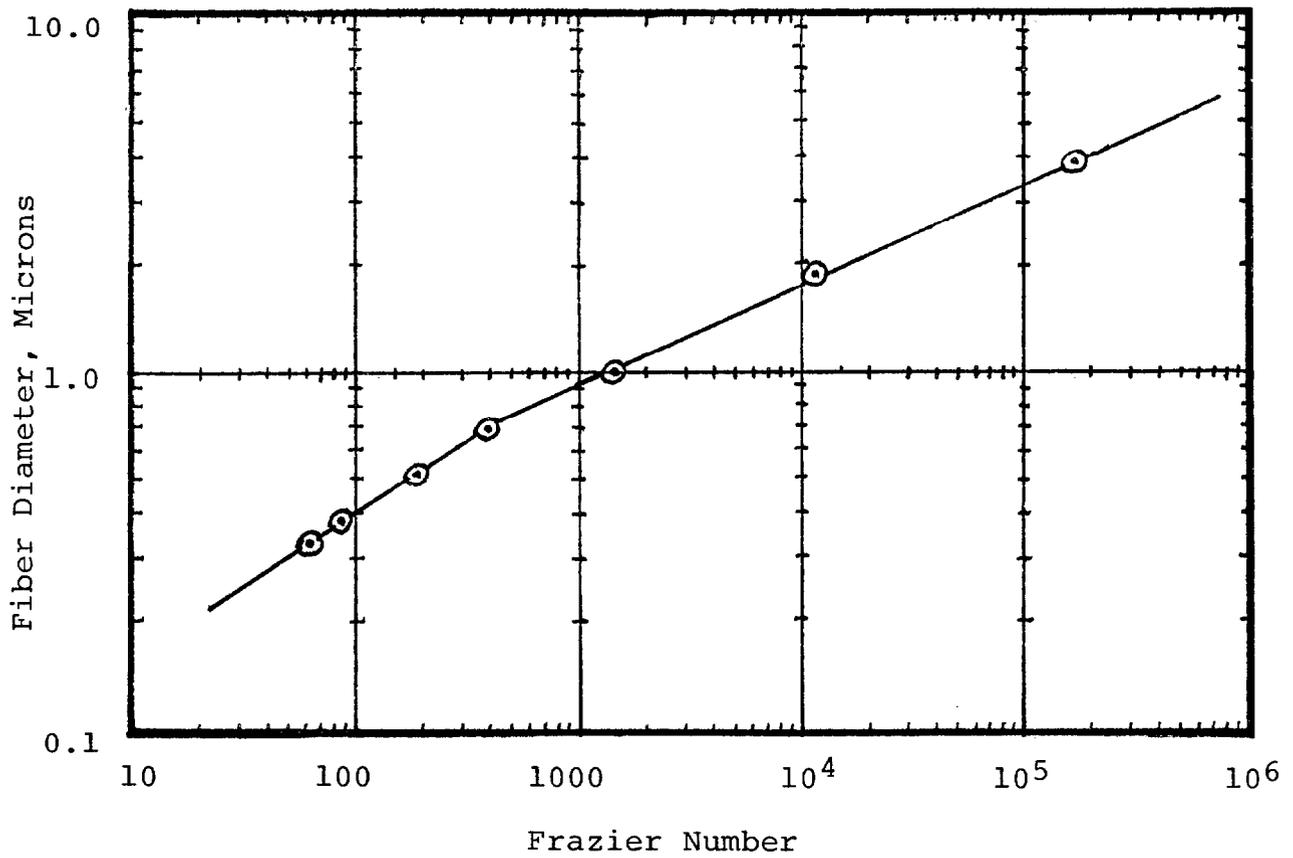


Figure III Frazier number versus fiber diameter



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EVALUATION OF DATA FROM HEPA FILTER QUALITY ASSURANCE TESTING STATIONS*

by

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U. S. Nuclear Regulatory Commission

In Revision 1 to Regulatory Guide 1.52, issued in July 1976, the NRC recommended that high efficiency particulate air (HEPA) filters for use in engineered safety feature (ESF) atmosphere cleanup systems be visually inspected and dioctyl phthalate (DOP) tested at either of two Department of Energy (DOE) operated QA Filter Testing Stations prior to their installation and use in commercial nuclear power plants. This recommendation was predicated on the practice initiated by DOE and its predecessor, the Atomic Energy Commission (AEC), in the early 1960's, and remains in effect today for all HEPA filters designated for use in DOE facilities. This practice was initiated because filter vendors were unable to consistently provide a HEPA filter that would meet the stringent requirements established by DOE and NRC and its predecessor the AEC.

In 1977, the NRC staff undertook a program to revise Regulatory Guide 1.52 to reflect recently issued industry standards (e.g., ANSI N509 and N510) and current industry practices. Revision 2 to Regulatory Guide 1.52 was formally issued in March 1978. In conducting this review, we reevaluated the recommendation that HEPA filters, intended for use in ESF systems in commercial nuclear power plants, be routinely tested at the DOE-QA Filter Testing Stations. As part of this evaluation we conducted a detailed analysis of the filter test results recorded by the two QA Testing Stations during the period 1971 to 1977. This paper will summarize the results of our analysis and explain the rationale for deleting the requirement that all HEPA filters intended for use in ESF systems be tested at the QA Testing Station prior to installation in nuclear power plants. Revision 2 to Regulatory Guide 1.52 (Section 3.d) recommends that HEPA filters be tested for penetration of DOP in accordance with the provisions of MIL-F-51068 and MIL-STD-282. In addition, it stipulates that the quality assurance requirements of Appendix B, "Quality Assurance Criteria for Nuclear Power Plants and Fuel Reprocessing Plants," to 10 CFR Part 50, be applied to all activities affecting the safety-related functions of HEPA filters. In effect, this revision places greater reliance on the filter vendors who already have the capability to construct HEPA filters in accordance with the requirements of ANSI N509 and to test their filters in accordance with the military specifications noted above.

During the period 1971 to 1977, the Oak Ridge and Hanford QA Testing Stations tested over 88,000 HEPA filters of all sizes and of this number, on the average, approximately 6% were rejected. Table 1 shows the number of filters tested of the four principal filter manufacturers, the number and percent of filters rejected, and the causes for these rejections. Of the filters tested, the principle cause of rejection was the failure of the filter to meet the penetration requirements. The number rejected because of excessive penetration ranged from a low of 16% to a high of 47%. What is not known, but would be most helpful, would be the measured penetration value compared to the acceptance criteria. This would permit an assessment of how far the penetration values exceed the acceptance criteria. To our knowledge, no error bar has been established for these tests, so the absolute value

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is imposed as the criteria for acceptance or rejection. Perhaps this is something that should be considered, since there certainly must be some error associated with the calibration and operation of the penetrometers.

As shown in Table 2, of the 88,000 filters tested, 30,966 filters had a rated capacity of 1000 cfm and less than 4% were rejected (1,167). Of these filters, less than 1,600 were specifically intended for use in ESF systems. Table 3 shows the number of filters tested from the four principal vendors, the number and percent of filters rejected, and the causes of these rejections. Of the 1,600 filters tested, approximately 1.5% were rejected due to failure to pass the penetration test or because of defective medium.

Table 4 shows the number of all types of filters tested at either DOE-QA Testing Stations, the percent rejected and the causes of these rejections. It is interesting to note that of all the filters tested, the 1000 cfm filters performed better than other filters tested (3.8% vs 6.4%).

Table 5 shows the total number of filters tested at Hanford for the four principal vendors, while Table 6 shows the results of 1000 cfm filters tested at this facility. Table 7 shows the total number of filters tested at Oak Ridge for the four principal vendors, while Table 8 shows the results of the 1000 cfm filters tested at this facility. Table 9, 10, 11, and 12 are included to show the semi-annual totals of the four filter manufacturers. For the period January 1, 1971 to September 30, 1977, rejection rates ranged from a low of 2% for vendor D to a high of 5.3% for vendor A for the filters tested at Oak Ridge, and from a low of 7.3% for vendor C to a high of 13.8% for vendor A for filters tested at Hanford. The reason for the higher rejection rate for filters tested at Hanford is not known. During this reporting period, Oak Ridge tested approximately 42,000 filters, while Hanford tested approximately 46,000. Since both facilities tested essentially the same number of filters, one could speculate that perhaps Hanford has a more rigorous inspection program or that there may be a difference in the acceptance criteria used by each facility. Whatever the reason, the wide difference should be determined.

In our opinion, the results of this evaluation show that the mandatory confirmatory tests at the DOE-QA Filter Testing Stations are not required for filters intended for use in ESF systems in nuclear power plants. We plan to monitor the test results from these facilities on a periodic basis to look for trends that may indicate a reversal in the presently low rejection rates.

In addition to the testing results, we also considered the cost to the industry of having these filters inspected at the QA Testing Stations prior to use in commercial nuclear power plants. For a typical powerplant, we considered three ESF systems (e.g., BWR-standby gas treatment system, fuel handling system, and control room system) each having a rated capacity of 10,000 cfm and each system redundant. We considered that the cost of the filters are increased by approximately 30% (\$125 to \$160). Assuming once-a-year change out of these redundant systems (60 filters) each reactor could realize a savings of \$2,100 or \$63,000 over the expected 30 year operating life of the plant. Equivalently, for a projected 200 operating reactors in 1985, a savings of \$420,000 per year to the nuclear industry can be predicted. Based on the small number of filters rejected as shown previously, the additional cost associated with testing of these filters to assure they satisfy MIL-51068D before installation is questionable. As a result of this evaluation, the NRC has deleted the requirement that all filters be tested at a DOE-QA Filter Testing Station prior to use in ESF systems installed in nuclear power plants. We would like to emphasize that this revision affects only NRC licensed nuclear power reactors and does not have an impact on any DOE facilities.

TABLE 1 - All Filters Tested At
 Either DOE Filter Test Facility

MANUFACTURER	NUMBER TESTED	NUMBER REJECTED (% REJECTED)	NUMBER REJECTED FOR EACH REJECTION CAUSE					
			PENETRATION	MEDIUM	CARRIER DAMAGE	FRAME	GASKET	MISCELLANEOUS
A	13068	1594 (12.2%)	264	249	76	617	254	134
B	10973	636 (5.8%)	258	107	41	47	106	77
C	46701	2462 (5.3%)	1121	432	59	193	418	239
D	17536	1089 (6.2%)	132	499	72	165	21	200
TOTAL	88278	5781 (6.5%)	1775	1287	248	1022	799	643

TABLE 2 - Filters of 1000 CFM Capacity Tested At Either DOE Filter Test Facility

MANUFACTURER	NUMBER TESTED	NUMBER REJECTED (% REJECTED)	NUMBER REJECTED FOR EACH REJECTION CAUSE						
			PENETRATION	MEDIUM	CARRIER DAMAGE	FRAME	GASKET	MISCELLANEOUS	
A	3573	371 (10.4%)	106	72	38	45	44	66	
B	1920	105 (5.5%)	69	12	17	2	1	4	
C	18061	424 (2.3%)	191	80	21	66	9	57	
D	7412	267 (3.6%)	5	112	14	29	3	104	
TOTAL	30966	1167 (3.8%)	371	276	90	142	57	231	

TABLE 3 - Filters of Capacity Greater Than or Equal to 1000 cfm for Nuclear Applications Tested at Either DOE Filter Test Facility

MANUFACTURER	NUMBER TESTED	NUMBER REJECTED (% REJECTED)	NUMBER REJECTED FOR EACH REJECTION CAUSE						
			PENETRATION	MEDIUM	CARRIER DAMAGE	FRAME	GASKET	MISCELLANEOUS	
A	133	0	0	0	0	0	0	0	0
B	21	1 (4.8%)	0	0	0	0	0	0	1
C	1053	22 (2.1%)	12	10	0	0	0	0	0
D	383	1 (0.3%)	1	0	0	0	0	0	0
TOTAL	1590	24 (1.5%)	13	10	0	0	0	0	1

TABLE 4 - All Filters Tested At
 Either DOE Filter Test Facility
 Differentiated by Filter Category

FILTER CATEGORY	NUMBER TESTED	NUMBER REJECTED (% REJECTED)	NUMBER REJECTED FOR EACH REJECTION CAUSE						
			PENETRATION	MEDIUM	CARRIER DAMAGE	FRAME	GASKET	MISCELLANEOUS	
ALL	85050	5472 (6.4%)	1715	1209	246	974	685	643	
1000 CFM	30966	1167 (3.8%)	371	276	90	142	57	231	
≠ 1000 CFM	23375	1183 (5.1%)	628	219	47	25	34	230	
≥ 1000 CFM	36620	1506 (4.1%)	524	316	103	141	125	297	
≥ 1000 CFM NUCLEAR	1590	24 (1.5%)	13	10				1	

TABLE 5 - ALL FILTERS TESTED AT
MANFORD

MANUFACTURER	NUMBER TESTED	NUMBER REJECTED (% REJECTED)	NUMBER REJECTED FOR EACH REJECTION CAUSE						
			PENETRATION	MEDIUM	CARRIER DAMAGE	FRAME	GASKET	MISCELLANEOUS	
A	10632	1464 (13.8%)	192	237	74	617	252	92	
B	2417	270 (11.2%)	101	60	13	37	54	5	
C	22841	1659 (7.3%)	552	385	54	190	400	78	
D	10442	950 (9.1%)	127	378	65	164	21	195	
TOTAL	46332	4343 (9.4%)	972	1060	206	1008	727	370	

TABLE 6 - Filters of 1000 CFM Capacity Tested At Hanford

MANUFACTURER	NUMBER TESTED	NUMBER REJECTED (% REJECTED)	NUMBER REJECTED FOR EACH REJECTION CAUSE						
			PENETRATION	MEDIUM	CARRIER DAMAGE	FRAME	GASKET	MISCELLANEOUS	
A	1983	292 (14.7%)	64	63	37	45	44	39	
B	273	36 (13.2%)	16	5	12	2	1	0	
C	3492	225 (6.4%)	50	61	18	63	6	27	
D	2973	253 (8.5%)	4	103	12	28	3	103	
TOTAL	8720	806 (9.2%)	134	232	79	138	54	169	

TABLE 7 - All Filters Tested At
Oak Ridge

MANUFACTURER	NUMBER TESTED	NUMBER REJECTED (% REJECTED)	NUMBER REJECTED FOR EACH REJECTION CAUSE						
			PENETRATION	MEDIUM	CARRIER DAMAGE	FRAME	GASKET	MISCELLANEOUS	
A	2436	130 (5.3%)	72	12	2	0	2	42	
B	8556	366 (4.3%)	157	47	28	10	52	72	
C	23860	803 (3.4%)	569	47	5	3	18	161	
D	7094	139 (2.0%)	5	121	7	1	0	5	
TOTAL	41946	1438 (3.4%)	803	227	42	14	72	280	

TABLE 8 - Filters of 1000 CFM Capacity Tested at Oak Ridge

MANUFACTURER	NUMBER TESTED	NUMBER REJECTED (% REJECTED)	NUMBER REJECTED FOR EACH REJECTION CAUSE						
			PENETRATION	MEDIUM	CARRIER DAMAGE	FRAME	GASKET	MISCELLANEOUS	
A	1590	79 (5.0%)	42	9	1	0	0	0	27
B	1648	69 (4.2%)	53	7	5	0	0	0	4
C	14569	199 (1.4%)	141	19	3	3	3	3	30
D	4439	14 (0.3%)	1	9	2	1	0	0	1
TOTAL	22246	361 (1.6%)	237	44	11	4	3	3	62

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TABLE 9 - Filters Manufactured By Vendor A. (Semi-Annual Totals)

DATE	OAK RIDGE			HANFORD			COMBINED		
	TESTED	REJECTED	% REJ.	TESTED	REJECTED	% REJ.	TESTED	REJECTED	% REJ.
Jan 1 to Jun 30, 1971	441	24	5.4%	287	45	15.7%	728	69	9.5%
Jul 1 to Dec 31, 1971	273	12	4.4%	262	23	8.8%	535	35	6.5%
Jan 1 to Jun 30, 1972	805	62	7.7%	861	115	13.4%	1666	177	10.6%
Jul 1 to Dec 31, 1972	659	19	2.9%	1210	137	11.3%	1869	156	8.3%
Jan 1 to Jun 30, 1973	69	8	11.6%	67	6	9.0%	136	14	10.3%
Jul 1 to Dec 31, 1973	1	0		327	69	21.1%	328	69	21.0%
Jan 1 to Jun 30, 1974	2	0		263	23	8.7%	265	23	8.7%
Jul 1 to Dec 31, 1974	0	0		148	33	22.3%	148	33	22.3%
Jan 1 to Jun 30, 1975	15	2	13.3%	72	3	4.2%	87	5	5.7%
Jul 1 to Dec 31, 1975	4	0		2110	281	13.3%	2114	281	13.3%
Jan 1 to Jun 30, 1976	84	0		2776	499	18.0%	2860	499	17.4%
Jul 1 to Dec 31, 1976	83	3	3.6%	1349	181	13.4%	1432	184	12.8%
Jan 1 to Sep 30, 1977	-	-	-	899	49	5.4%	900	49	5.4%
Total	2436	130	5.3%	10632	1464	13.8%	13068	1594	12.2%

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TABLE 10 - Filters Manufactured by Vendor B. (Semi-Annual Totals)

DATE	OAK RIDGE			HANFORD			COMBINED		
	TESTED	REJECTED	% REJ.	TESTED	REJECTED	% REJ.	TESTED	REJECTED	% REJ.
Jan 1 to Jun 30, 1971	229	21	9.2%	133	17	12.8%	362	38	10.5%
Jul 1 to Dec 31, 1971	550	60	10.9%	191	28	14.7%	741	88	11.9%
Jan 1 to Jun 30, 1972	245	35	14.3%	204	7	3.4%	449	42	9.4%
Jul 1 to Dec 31, 1972	514	21	4.1%	40	5	12.5%	554	26	4.7%
Jan 1 to Jun 30, 1973	439	30	6.8%	104	12	11.5%	543	42	7.7%
Jul 1 to Dec 31, 1973	803	64	8.0%	154	27	17.5%	957	91	9.5%
Jan 1 to Jun 30, 1974	370	8	2.2%	76	8	10.5%	446	16	3.6%
Jul 1 to Dec 31, 1974	1078	30	2.8%	8	0		1086	30	2.8%
Jan 1 to Jun 30, 1975	717	6	0.8%	31	4	12.9%	748	10	1.3%
Jul 1 to Dec 31, 1975	1190	6	0.5%	417	76	18.2%	1607	82	5.1%
Jan 1 to Jun 30, 1976	1174	11	0.9%	324	11	3.4%	1498	22	1.5%
Jul 1 to Dec 31, 1976	1247	74	5.9%	566	39	6.9%	1813	113	6.2%
Jan 1 to Sep 30, 1977	-	-	-	169	36	21.3%	169	36	21.3%
Total	8556	366	4.3%	2417	270	11.2%	10973	636	5.8%

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TABLE 11 - Filters Manufactured By Vendor C (Semi-Annual Totals)

DATE	OAK RIDGE			HANFORD			COMBINED		
	TESTED	REJECTED	% REJ.	TESTED	REJECTED	% REJ.	TESTED	REJECTED	% REJ.
Jan 1 to Jun 30, 1971	1765	10	0.6%	1032	44	4.3%	2797	54	1.9%
Jul 1 to Dec 31, 1971	3800	78	2.1%	1042	31	3.0%	4842	109	2.3%
Jan 1 to Jun 30, 1972	2654	78	2.9%	1547	119	7.7%	4201	197	4.7%
Jul 1 to Dec 31, 1972	1998	159	8.0%	1680	92	5.5%	3678	251	6.8%
Jan 1 to Jun 30, 1972	1651	119	7.2%	1773	366	20.6%	3424	485	14.2%
Jul 1 to Dec 31, 1973	3022	110	3.6%	1055	129	12.2%	4077	239	5.9%
Jan 1 to Jun 30, 1974	1733	44	2.5%	2621	51	1.9%	4354	95	2.2%
Jul 1 to Dec 31, 1974	603	11	1.8%	3157	145	4.6%	3760	156	4.1%
Jan 1 to Jun 30, 1975	309	3	1.0%	1608	89	5.5%	1917	92	4.8%
Jul 1 to Dec 31, 1975	1672	46	2.8%	1327	91	6.9%	2999	137	4.6%
Jan 1 to Jun 30, 1976	1573	32	2.0%	2735	110	4.0%	4308	142	3.3%
Jul 1 to Dec 31, 1976	3080	113	3.7%	1357	190	14.0%	4437	303	6.8%
Jan 1 to Sep 30, 1977	-	-	-	1907	202	10.6%	1907	202	10.6%
Total	23860	803	3.4%	22841	1659	7.3%	46701	2462	5.3%

TABLE 12 - Filters Manufactured by Vendor D (Semi-Annual Totals)

DATE	OAK RIDGE			HANFORD			COMBINED		
	TESTED	REJECTED	% REJ.	TESTED	REJECTED	% REJ.	TESTED	REJECTED	% REJ.
Jan 1 to Jun 30, 1971	1268	101	8.0%	38	0		1306	101	7.7%
Jul 1 to Dec 31, 1971	187	0		58	0		245	0	
Jan 1 to Jun 30, 1972	483	1	0.2%	14	0		497	1	0.2%
Jul 1 to Dec 31, 1972	144	5	3.5%	3796	292	7.7%	3940	297	7.5%
Jan 1 to Jun 30, 1973	462	12	2.6%	2226	222	10.0%	2688	234	8.7%
Jul 1 to Dec 31, 1973	498	3	0.6%	1565	170	10.9%	2063	173	8.4%
Jan 1 to Jun 30, 1974	625	3	0.5%	1156	37	3.2%	1781	40	2.2%
Jul 1 to Dec 31, 1974	799	1	0.1%	135	17	12.6%	934	18	1.9%
Jan 1 to Jun 30, 1975	1283	1	0.1%	304	21	6.9%	1587	22	1.4%
Jul 1 to Dec 31, 1975	393	2	0.5%	211	18	8.5%	604	20	3.3%
Jan 1 to Jun 30, 1976	454	0		478	65	13.6%	932	65	7.0%
Jul 1 to Dec 31, 1976	498	10	2.0%	209	86	41.1%	707	96	13.6%
Jan 1 to Sep 30, 1977	-	-	-	252	22	8.7%	252	22	8.7%
Total	7094	139	2.0%	10442	950	9.1%	17536	1089	6.2%

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DISCUSSION

CADWELL: Any attempt to save money and streamline operations is appreciated. My interpretation of your data is that the Quality Assurance Stations have done an effective job in keeping the quality of the filters high. Our fear is that, if you take away the threat of rejection at the retest stations, there will be a gradual deterioration of the quality of HEPA filters with a resultant deterioration in the reactor safety program.

COLLINS: I have to take issue with you. I don't believe that the Quality Assurance Station by itself has been the primary reason for continuing good performance of the filters. I think that the nuclear industry has grown to a point where it realizes the need to produce a product comparable to other components of the nuclear power plant. I think the initiative and the efforts of the ANSI standards people and the industry itself to regulate the quality of filters have been the principal reasons, rather than the Quality Assurance Stations.

CADWELL: I agree with that but I am afraid that if you take away the threat of financial loss due to rejection at the Stations you open the door of compromise between the manufacturer and the buyer. That could become a potentially dangerous situation.

COLLINS: That could be a result but, as I indicated, we certainly are going to continue to monitor the program. Nevertheless, I think that the HEPA filter is only one component in the total nuclear power plant and if you take the kind of approach you are advocating, it could be extended to all other components, particularly to those in the pressure boundary systems. If we started to impose the practice of Quality Assurance Stations for all other components, I believe the nuclear industry would be on our back. I believe that we, as regulators, have a responsibility, when we see data that show trends, to indicate that testing is no longer required for government installations, i.e., that the industry can regulate itself, to "de-ratchet" not to "ratchet" for higher controls.

MURROW: Information on manufacturers A through D is impressively good but manufacturers "E" through "X" have no existence and no past history. How can we be assured they will produce as good filters when they enter the market?

COLLINS: I can appreciate that; the data that were available to us showed only the four principal vendors. The only answer I can give is that, if the rest of the alphabet comes along and if they can meet the requirements of Appendix B and can test filters in accordance with the ANSI standards and the Mil-Specs, more power to them. They certainly would be acceptable.

BURCHSTED: I would like to make five statements. (1) Past is not prelude; quality generated by the industry in the past has no bearing on quality of non-monitored filters in the future. (2) There is a fundamental quality assurance principle that the workman does not inspect his own work. (3) The proposed monitoring of the filter test stations has little bearing on filters supplied to the commercial power industry which will no longer require confirmatory quality assurance testing. (4) It is possible that abandonment of confirmatory test-station testing may lead to the development of two grades of filters, those subject to confirmatory testing, and those not subject to such testing. It is significant that Mr. Cadwell, a filter manufacturer, apparently believes the same. (5) As a very minimum, I would suggest that the Nuclear Regulatory Commission should obtain occasional samples of the filters furnished to power stations and have them tested by the filter test stations.

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COLLINS: I cannot argue with some of your statements but if you would realize the number of filters intended for use in commercial nuclear power plants was 1600 and that of that number only 1.5% were rejected, that is pretty good quality.

BURCHSTED: I accept that, but just because this happened in the past, it doesn't mean it will happen in the future in the absence of tests by Quality Assurance Stations.

COLLINS: That is very true. I think the only thing that we can say is that, in my opinion and in the opinion of the staff, the past is something that we should pay attention to. The experience we have gained is something that we must use. If it turns out that we were wrong, we were wrong, and it won't be the first time, but we try not to let it happen often.

KOVACH: I have two comments. One is that we at NUCON have not seen a difference between failure of installed components which were or were not tested at a QA station. Our observation is that damage to, or the failure of, HEPA filter systems is much more often caused by sloppy installation than a failure caused by poor filter manufacture or by excess shipping or by QA testing of the filters. This confirms your conclusions.

At the same time, some periodical and random checks on the manufacturers would be very helpful to assure maintenance of the quality of a fragile component of ESF and other systems. Maybe this is aiming at a compromise, but I believe it may be advantageous for the users to keep track of the manufacturers.

COLLINS: I appreciate the comment. What you have recommended has been proposed to us and I am giving it very serious consideration.

DAVIS: Will there be a continued requirement for in-place testing of HEPA filter systems at power plants, and is this a factor in the decision to discontinue the requirement for QA testing at the DOE test station?

COLLINS: Oh, yes! We would never eliminate the requirement for in-place testing. In-place test requirements are imposed on a licensed facility by its operating license as part of the technical specifications.

DAVIS: Is that a part of your decision then?

COLLINS: No, that has always been in the technical specifications and the tests will continue.

DAVIS: I mean, is that a part of the decision to dispense with the test station requirement.

COLLINS: It certainly had an impact on our decision.

FRY: Can you tell me what the frequency of filter sampling was? The reason I ask is that the situation you describe is a classic QC (quality control) problem. The quality of the product is good at the current frequency of sampling. In such a case, one simply reduces the frequency of sampling until a problem is identified and then the frequency of sampling is increased again. This is, in fact, exactly what you are doing by continuing to "monitor" product quality.

COLLINS: That is correct.

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STEINBERG: The arbitrary decision to cease QA testing of filters and subsequent closing of the QA Stations is a giant leap backward. Now, the filter suppliers will take advantage of the relaxed requirements and the possibility of installation of filters with defects will inevitably increase.

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PERFORMANCE AND ENVIRONMENTAL CHARACTERISTICS OF A COMPACT, HIGH-CAPACITY HEPA FILTER DESIGN

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Abstract

An improved version of the HEPA filter has been developed and evaluated for conformity with MIL-F-51068. In this design, the filter paper (conforming to MIL-F-51079) is pleated to a depth of only 18 mm. Ribbons essentially the same material as the filter paper form inter-pleat spacers, providing a crest-to-crest spacing of 3 mm. The resultant filter has the same DOP penetration level as the standard MIL-F-51068 HEPA filter, but about half its pressure drop at the same airflow. Materials of construction, adhesives, gaskets and faceguards match MIL-F-51068. Both flat-panel and zig-zag configurations of the filter meet the rough handling test of MIL-F-51068. Preliminary indications are that both filters will meet the overpressure tests of MIL-F-51068D and the flammability requirements of UL 586; these tests are in progress. Filters can be supplied to interchange with existing HEPA filter sizes. The improved pressure drop characteristic of these filters can be used to provide longer intervals between filter changes, or equal life at greater capacity.

I. Introduction

The basic design of HEPA filters used in American nuclear facilities has changed little since the original development of this filter during the 1940's. In this design, a web of paper is formed into pleats of depth from about 70 mm to about 300 mm, depending on the desired gas flow velocity through the completed filter. These pleats are held apart both upstream and downstream by corrugated spacers which provide a pleat pitch of about 9 mm. This media-separator pack is sealed into wood or metal cell sides to form a filter cartridge. The design has proved to be effective and reliable in a wide range of environmental conditions, when the proper component materials have been used. Component materials for both military and civilian nuclear applications are defined in MIL-F-0051079 (for the filter media) and MIL-F-0051068 (for the completed filter cartridge).

These specifications are definitive enough to give the impression that only minor deviations from the standard design are possible. The requirements imposed by MIL-F-0051079 on the filter media for example, essentially dictate a wet-laid glassfiber paper with a furnish of fibers predominantly less than 1 μ m in diameter. MIL-F-0051068 is even more restrictive in defining the design of the completed filter, since it specifies many construction details, as well as performance requirements. In both the "D" and "E" versions of this specification, however, wording exists allowing some material and construction deviations from the specified norms. Section 3.2.6, for example, permits the procuring agency to specify spacing means

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different from those stated in MIL-F-0051068. Filters made with such deviations must, of course, meet the environmental and performance requirements of the specification to be acceptable for nuclear service. One U.S. manufacturer of HEPA filters does produce a filter which has no separators at all, relying on a special double-pleating technique to provide the necessary air passages; this filter is accepted for nuclear applications. Thus it appears that deviations from MIL-F-0051068 can be permitted in U.S. nuclear applications, if reliability can be maintained, and if some advantage is obtained by the change.

A form of high efficiency particulate filter quite different from the U.S. design has been in use in Europe for several years. Figure 1 shows its design. Here the filter media is not pleated to the full depth of the filter cartridge in the direction of air flow. Instead, pleats are no more than 20 mm deep, and the crest-to-crest pitch is about 3 mm. Adjacent pleats are separated by ribbons of foam plastic, asbestos, or glued-on threads. A full size filter cartridge is fabricated from several panels of this construction arranged in a zig-zag fashion. This design allows considerably more area of filter media to be exposed to air flow in a given filter cartridge than is possible with the U.S. design. Such increased area provides a lower pressure drop at increased flow, or extended service life at normal flow.

Earlier European designs, like their American counterparts, did not pay great attention to flammability, radiation and humidity resistance. However, more recent versions have incorporated glassfiber filter media meeting MIL-F-0051079, nonflammable spacer strips, and adhesives of greater radiation resistance.

It appears entirely possible to qualify a filter of this European design under MIL-F-0051068. The discussion which follows describes the results of MIL-F-0051068-type tests on a European-style HEPA filter of U.S. fabrication. In addition, data on resistance versus air flow at various dust loadings are given. These tests indicate that the European filter design can equal the American design in environmental resistance, and offers substantially longer life for the same cartridge dimensions and pressure drop.

II. Filter Description

The filters tested have been of two forms: flat panels of pleated media (Fig. 2) and assemblies of such panels (Fig. 1). In both cases, the filter media was the same, with the following characteristics:

Media furnish: glassfiber, average fiber diameter $< 1 \mu\text{m}$
Caliper: 0.015 in (0.38 mm) minimum
Basis Weight: 55 lb/3000 ft² (89.5 g/m²)
Flat Sheet air flow resistance: 40 mm WG (392 Pa) at 320 cm/min
Dry Tensile Strengths (Typical)

Machine direction, uncreased:	5.0 lb/in (8.76 N/cm)
Machine direction, creased :	2.0 lb/in (3.50 N/cm)
Cross direction, uncreased :	3.8 lb/in (6.65 N/cm)

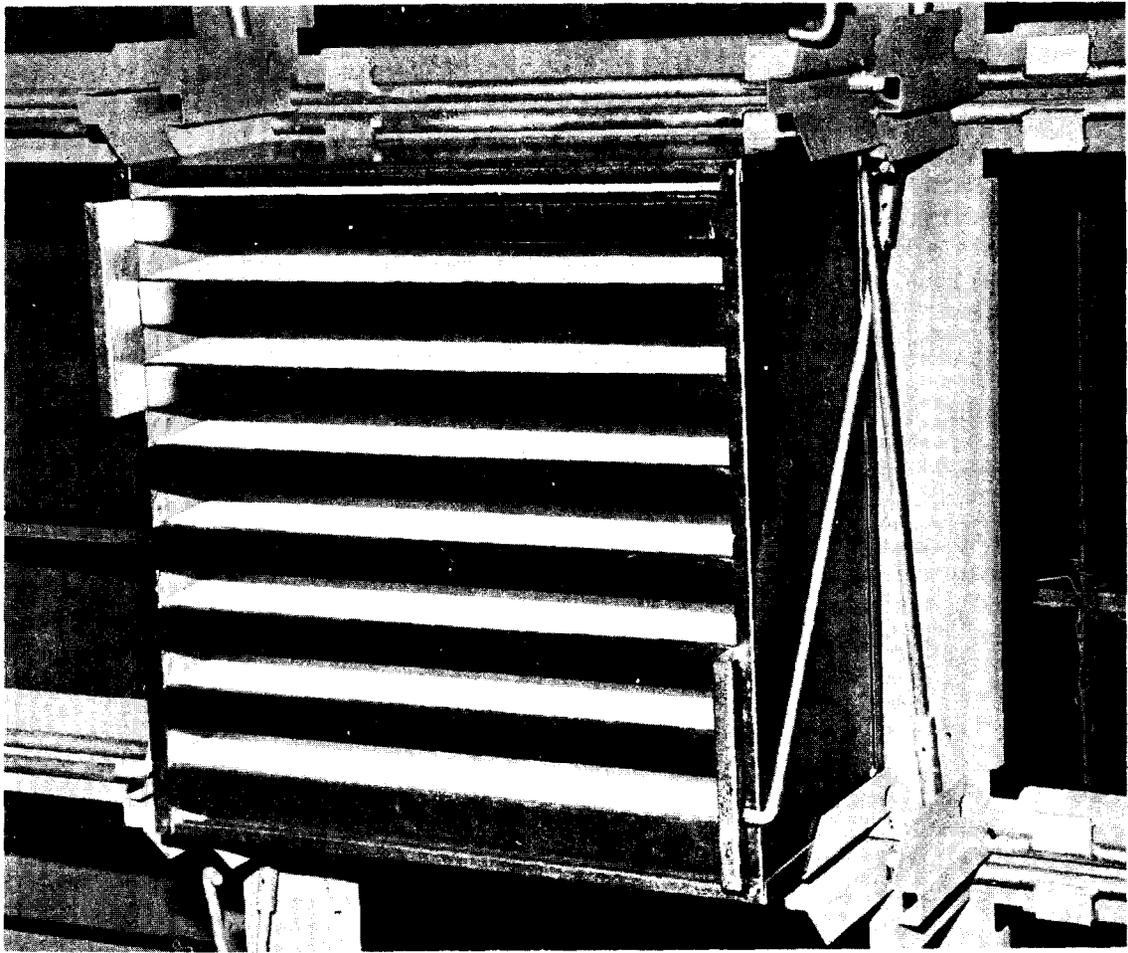


Figure 1. Close-Pleat Multipanel Filter with Quick-Release Clamps

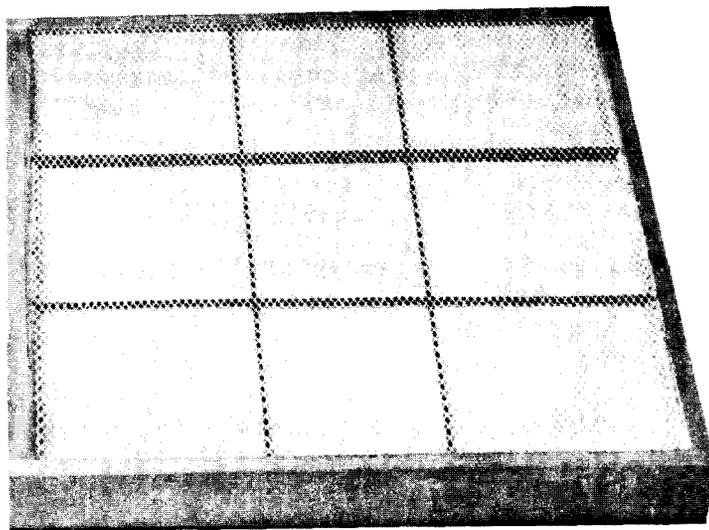


Figure 2. Close-Pleat Flat Panel Filter with Faceguard and Stiffeners

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Elongation at rupture, typical: 3.3%
DOP Penetration (Q127-5 Penetrometer, 0.3 μm diameter smoke,
320 cm/min, typical): 0.015%

This filter media has been in use for several years for conventional American HEPA filter production, and is fully qualified under MIL-F-0051079B.

For the flat panel filter, the peak-to-peak pleat depth is 50 mm; for panels used in the assembly of the zig-zag multipanel filter, this depth is 18 mm. Spacer ribbons are slit from rolls of glassfiber media very similar to the filter media itself. These ribbons are 1/8 in (3.2 mm) wide and of a caliper which yields a crest-to-crest pitch of 3.27 mm on the flat panel filters and 2.68 mm on the zig-zag multipanel filters. The center-to-center distance for the spacer ribbons is 31.5 mm.

A comparison between total media areas for this form of filter and for those listed in MIL-F-0051068D is interesting:

Filter Size	Dimensions mm	Media Area, m^2/filter	
		MIL-F-0051068D	Astrocel II and III
1	203x203x78	0.54	0.82 (1)
5,6	610x610x292	19.16	35.39 (2)

Notes: (1) Astrocel II, flat panel (2) Astrocel III, zig-zag multipanel

The spacer ribbons are held in place by friction, rather than by an adhesive. Thus airflow is not entirely cut off from the media area covered by the ribbons. In addition, the flammability and environmental resistance of the spacer material is essentially identical to the filter medium itself. (See Figs. 3 and 4 for construction details.)

As in the case of conventional HEPA filter design, cell sides can be made of various materials. For the flat panel filters, particle board, plywood, galvanized sheet steel and aluminum extrusions have all been used. The design described here is currently available only in plywood. For the zig-zag multipanel filter, European practice is to form cell sides from stainless, galvanized or painted sheet steel. The design described here is (at this writing) available in 20 ga. galvanized sheet steel only.

Several types of sealant have been used by European manufacturers to seal the edges of the packs to the cell sides: bitumen, silicones, polyvinyl chloride, and other synthetic resins and plastisols. The design described here is sealed with a two-part epoxy containing a fire retardant.

Faceguards, as shown in Fig. 2, are essential to the flat panel filter design for this service. The zig-zag design, however, is inherently protected by the channels which span the face of the filter and seal the upstream and downstream edges of the media panels. Expanded metal faceguards can be added to this filter also, of course.

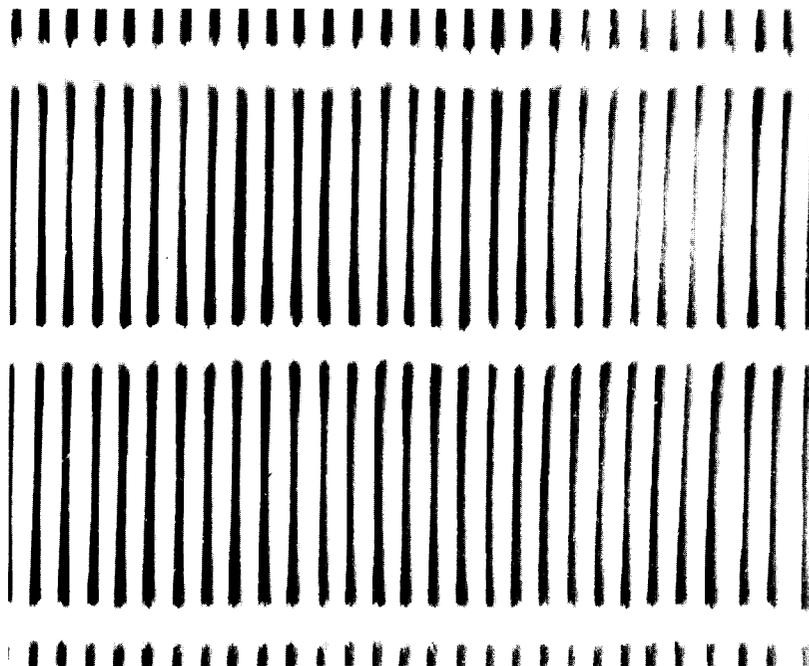


Figure 3. Close-Pleat Filter Media Pack with Spacer Ribbons

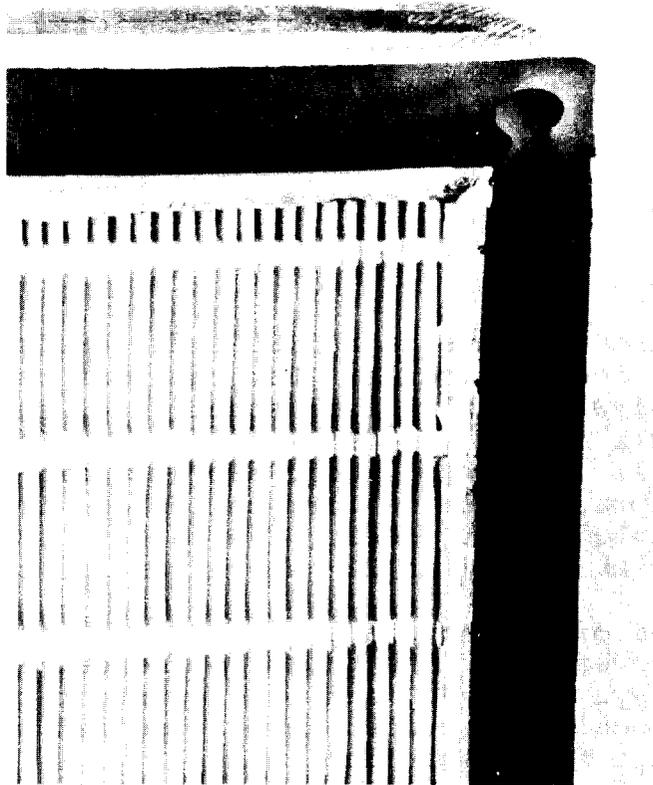


Figure 4. Sealant and Dovetail Gasket Joint, Flat Panel Filter

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HEPA filters with wood cell sizes are able to withstand clamping forces applied through the body of the cell side with little difficulty. Rather heavy gage steel cell sides are needed to match the compression resistance of wood. For this reason, the present design is preferably clamped at four points around the gasketed face adjacent to the support frame. Small oval cutouts through the cell sides allow clamping dogs to swing into position on the back side of the clamping flange. These clamping dogs are attached to actuator arms which allow them to be rotated out of clamping position very quickly. These arms also act as torsion bars to maintain gasket pressure as time passes (See Fig. 1). (A detailed description of this quick-release clamping arrangement is contained in Ref. 1). Clamping by more conventional means, with clamping force applied to the filter cell sides along the edges which are opposite the sealing gasket, is entirely practical, but less reliable than clamping by dogs at the sealing face. Thus filters of this design are interchangeable with standard American HEPA designs. Gaskets are SCE-43, in accordance with MIL-F-0051068D. Plywood cell sides for flat panel filters are fire retardant, again in accord with MIL-F-0051068D. Adhesives used contain fire retardants, and are self-extinguishing.

III. Performance Tests

The DOP penetration for both flat panel and multipanel filters, measured with the usual Q107 thermal DOP generator producing $0.3 \mu\text{m}$ particles is typically 0.01%, well below the 0.03% specified in MIL-F-0051068D.

Fig. 5 plots resistance of the zig-zag multipanel design at various dust loadings and air flows. Comparison data for the standard HEPA filter design are also shown. The dust fed was ASHRAE Standard Air Cleaner Test Dust minus its lint component. The standard design appears to build up resistance about three times as quickly as the zig-zag multipanel design.

Fig. 6 compares the resistance build-up for $500 \text{ ft}^3/\text{min}$ ($850 \text{ m}^3/\text{h}$) filters, the standard unit being $610 \times 610 \times 149 \text{ mm}$, and the close pleat design being $610 \times 610 \times 78 \text{ mm}$. The two filters track fairly closely up to 400 g dust load, after which the standard filter rises somewhat more slowly. However, the resistance at 400 g load was 3 in WG (747 Pa) which is quite high for normal service for this type of filter.

The smaller volume of the close-pleat panel filter, and its lesser weight (3.6 kg vs 9 kg for the standard design) undoubtedly reduce the waste disposal problem for this size unit, even if the two have equal lives.

IV. Environmental Tests

To date, we have been able to run only two of the environmental exposure tests specified in MIL-F-0051068D. The large capacity of the zig-zag multipanel unit means that a very large fan is needed to raise its resistance to 10 in WG (2490 Pa) as specified in MIL-F-0051068D. This test was simulated for a single slot of the

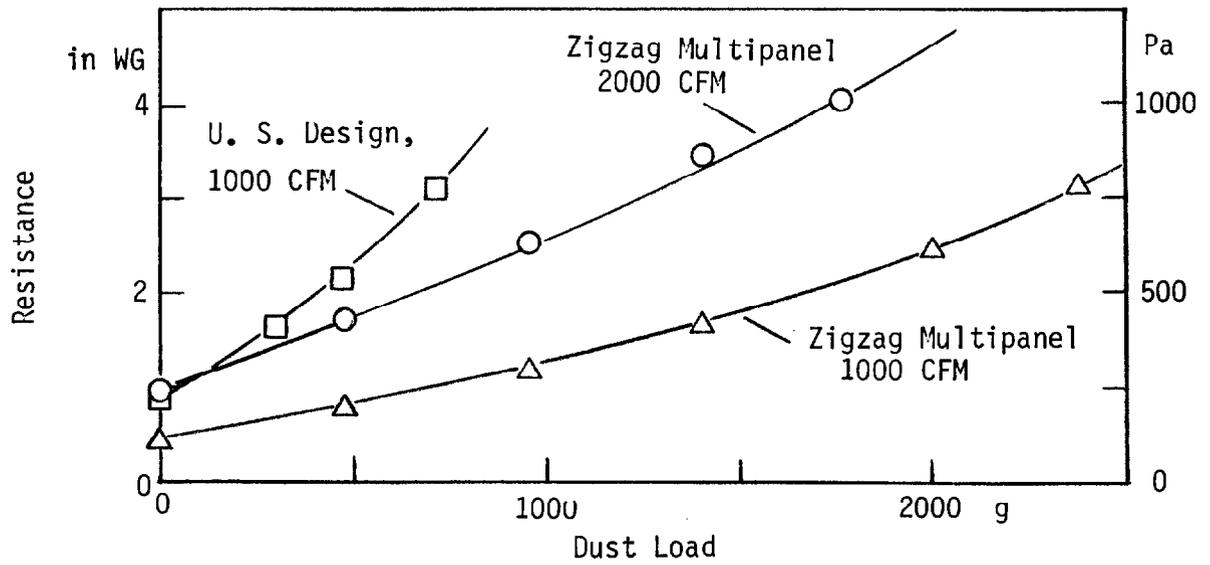


Figure 5. Resistance vs Dust Load for 610x610x292 mm Filters

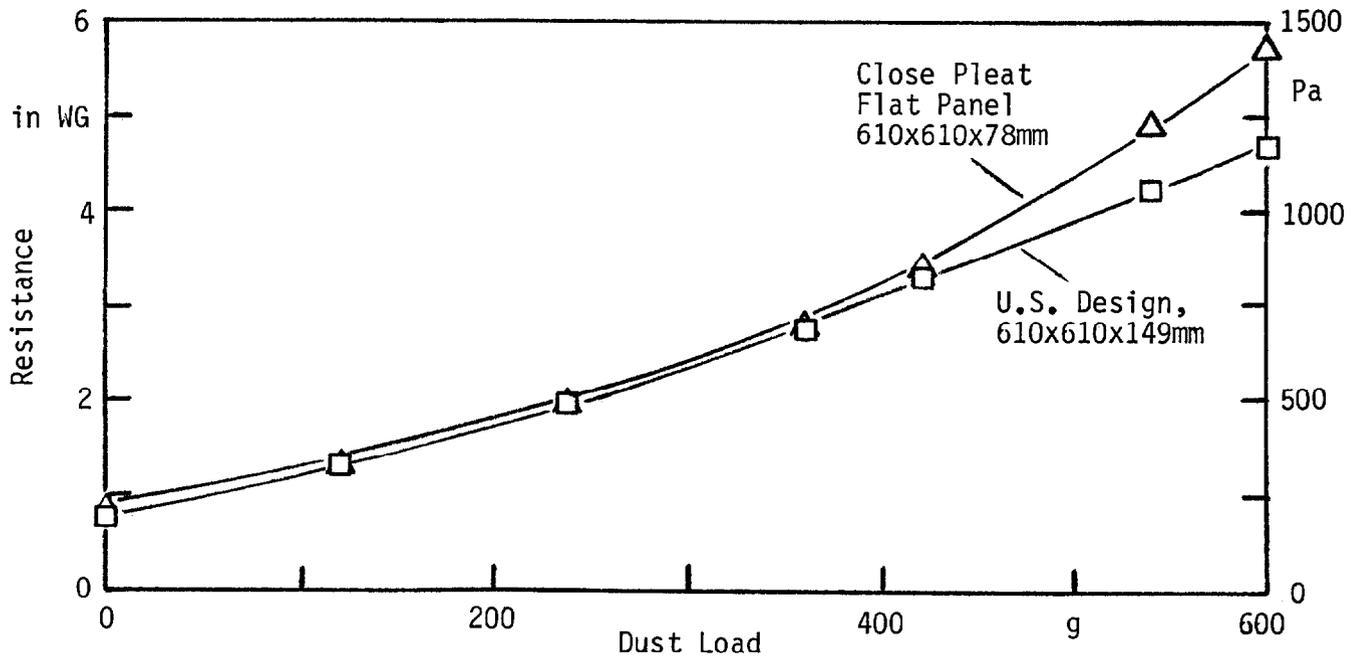


Figure 6. Resistance vs Dust Load for 500 ft³/min Filters

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zig-zag structure, and it was found that some downstream support between panels is needed to withstand the full 2490 Pa pressure. The full exposure, with DOP and resistance tests run before and after the overpressure exposure, was run on a close pleat flat panel unit, with the results listed in Table 1. The inherent structure of the media panel, plus the expanded metal faceguard and added crossbars (See Fig. 2) is apparently sufficient to qualify this filter.

Table I. Overpressure Tests.

Astrocel II Flat Panel Close-Pleat Filter

Filter No.	DOP Penetration, %		Pressure Drop At Rated Flow, mm WG	
	Before Exposure	After Exposure	Before Exposure	After Exposure
11	0.004	0.004	22.6	23.6
12	0.006	0.008	23.4	24.4
13	0.004	0.002	22.9	24.4

Filter dimensions 610x610x70 mm

Rated Flow: 500 cfm (850 m³/hr)

Exposure: Pressure Drop increased to 2490 Pa by increasing air flow. This condition was held for 15 minutes before flow was returned to rated level, and DOP Penetration and rated flow pressure drop "After" determined.

The rough handling test of MIL-F-0051068D, using the Q110 Vibrating Machine, was run on a zig-zag multipanel filter, with results listed in Table 2. This unit appears to survive this exposure without loss of performance.

A program exposing both filters to the remaining conditions of MIL-F-0051068D is now under way, as is the U.L. 586 series of flammability tests. The results of these tests will be reported when they are completed.

Table II. Rough Handling Tests
on 610x610x298 mm
Close Pleat Zig-Zag Multipanel Filter.

	1000 ft ³ /min	200 ft ³ /min
DOP Penetration before rough handling	0.018%	0.017%
DOP Penetration after vertical rough handling	0.017	0.020
DOP Penetration after horizontal rough handling	0.013	0.015

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Filter tested for DOP penetration "encapsulated" at 1000 cfm and 200 cfm.

Test sequence:

1. Rough handled with metal channels vertical on Q-110 rough handler - 200 cycles per min, 3/4 in. amplitude. 15 min. test
2. DOP test
3. Rough handle same filter with metal channels horizontal.
4. DOP test

References:

1. Rivers, R.D. (ed.) Design and Testing of Fan-Cooler-Filter Systems for Nuclear Applications. (USAEC Topical Report AAF-TR-7101) 1972

DISCUSSION

CADWELL: What will be the availability and relative price of the multi-panel, zig-zag filter?

RIVERS: The flat ones are available in any quantity, the multi-panel and zig-zag ones in small quantities. Our line isn't up to full speed yet.

CADWELL: Do you have any idea of the price yet?

RIVERS: No, I do not.

BURCHSTED: At the time we published the Nuclear Air Cleaning Handbook (ERDA 76-21), none of the European-design filters had met the U.S. requirements for nuclear-grade filters. John Dymont also commented that we had not discussed European practice in his review of the Handbook in "Filtration and Separation." When we re-issue the Handbook we would like to remedy this shortcoming, and I invite the overseas participants at this Conference to take steps to inform me accordingly,

RIVERS: I hope by the time of the 3rd Edition that we will have American experience, as well.

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PERFORMANCE OF 1,000 AND 1800 CFM HEPA FILTERS ON LONG EXPOSURE TO LOW ATMOSPHERIC DUST LOADINGS*

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Abstract

Comparative tests are in progress to determine the performance characteristics of European-design HEPA filters compared to U.S.-design units made with the same filter paper. European filters are being operated at their rated capacity of 1,800 cfm and at 1,000 cfm. The U.S.-design unit is operated at 1,000 cfm. Filters, installed on the roof of a 15-story building, are continuously exposed to an atmospheric dust aerosol of low concentration and small particle size. Plans are to continue these tests over a period of two or more years. Early results confirm that 1,800 cfm rated filters will have a very slow rate of pressure increase when operated at 1,000 cfm and an extended service life.

Introduction

Recent rapid increases in the price of nuclear solid waste disposal services have made it considerably more costly to dispose of a used HEPA filter than to acquire and install it. As there appears to be little immediate hope that important increases will take place in disposal productivity, total filter disposal costs can be lowered only by reducing the number of units discarded. A way that this desirable objective can be achieved without reducing the number of filters in essential service is to make them last longer. Obvious ways to accomplish this are to continue to operate HEPA filters until they reach a higher airflow resistance before replacement or to install more efficient (and higher resistance) prefilters. However, for existing systems, installed blowers and blower motors will be unable to deliver the required airflow at the higher filter resistance. Although the higher resistance option is open to designers of new filter systems, governmental requirements for energy conservation in new construction make it less attractive than it might have been at an earlier time.

Continental European filters have differed substantially in construction details from American and British HEPA filters. For many years, these European filters failed to find favor in the U.S. because the paper that was used in them was considered to be inferior. This no longer seems to be correct and interest in European filters has been rekindled because their design makes it possible to put sufficient effective filter paper into the standard 24-in. x 24-in. x 11 1/2-in. filter frame to handle 1,800 - 2000 cfm (instead of 1,000 cfm) at a clean filter resistance of 1-in. w.g. or less and to meet the maximum DOP penetration standard of 0.03% at these higher volumetric flowrates. The increase in air handling capacity is made possible by folding the filter paper into small pleats with very narrow air spaces between adjacent folds, thereby packing much more filter paper into the assigned space than is possible with the Amer-

*Work performed under U.S. Department of Energy Contract No. EY-S-02-3049 by the Harvard Air Cleaning Laboratory; Mr. J. Dempsey, Division of Waste Management, Contract Administrator.

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ican and British filter design that uses bulky corrugated separators between large paper pleats. Flanders Filters, Inc. increased air-flow capacity of one of their HEPA filter products by going to a separatorless design (Super-Flow) but they still use relatively large pleats and have not been able to obtain as great an increase in air-flow capacity as have the European manufacturers who use smaller and shallower pleats that are separated from adjacent folds by glued-on thin strips of paper or braided string at approximately 1-inch intervals. Stacks of the shallow pleats are placed in the filter frame with the open ends faced perpendicular to the direction of upstream airflow, and with alternating inlet and outlet air channels extending into the filter frame between the stacks of pleated filter as shown in Figure 1. Alternate air channels are blocked on the upstream side and on the downstream side so that all air that enters must pass through the pleated paper to reach an exit air channel.

Because the resistance of HEPA filters is directly proportional to air flow rate, when an 1800 cfm rated filter is operated at 1000 cfm, its clean resistance will be only 55% of what it is at the higher flow rate (e.g., from 1.0 to 0.55 in. w.g.). On the assumption that an 1800 cfm rated filter has 1.8 times more effective filter paper surface than a 1,000 cfm rated filter, the dust accumulation on a unit area of paper of an 1800 cfm rated filter, when operated at 1,000 cfm, will be only 55% of the amount deposited when the filter is operated at its rated flow. This means that the airflow resistance buildup of a 24 x 24 x 11 1/2-in. 1800 cfm-rated filter, operated at 1,000 cfm, will be decreased proportionately by the lower unit dust deposit rate as well as by the lower airflow rate through both dust deposit and paper. Theoretically, the rate of resistance buildup will reflect the product of the two effects and in the case cited above, the rate of resistance increase should be only 0.3 as fast, i.e., $0.55 \times .55 = 0.30$. Put another way, when operated at 1,000 cfm, the period required for an 1800 cfm rated filter to reach its maximum rated resistance (usually 3-4 in. w.g.) will be three times as long and, as a result, the number of filters discarded to waste will be reduced by two thirds. This represents a very impressive potential saving in waste disposal costs should practise be shown to follow theory.

Accelerated life tests of HEPA filters have been conducted with simulated aerosols, such as NBS filter test dust, at particle concentrations several orders of magnitude greater than HEPA filters are ordinarily subjected to in nuclear service. These results have notoriously poor predictive value for filters exposed to low dust loadings containing particles of much smaller size. The disadvantage of performing filter life tests with realistic aerosol concentrations and particle sizes is the very long times that are needed to arrive at reliable conclusions. This is unfortunate, but unavoidable.

Test Arrangements

Atmospheric dust has been widely used for high efficiency filter testing but near ground level, particle size and concentration are still greater than HEPA filters used in nuclear service are likely to be exposed to. If, however, ambient air substantially above ground level is selected as the test aerosol, dust concentrations and dust

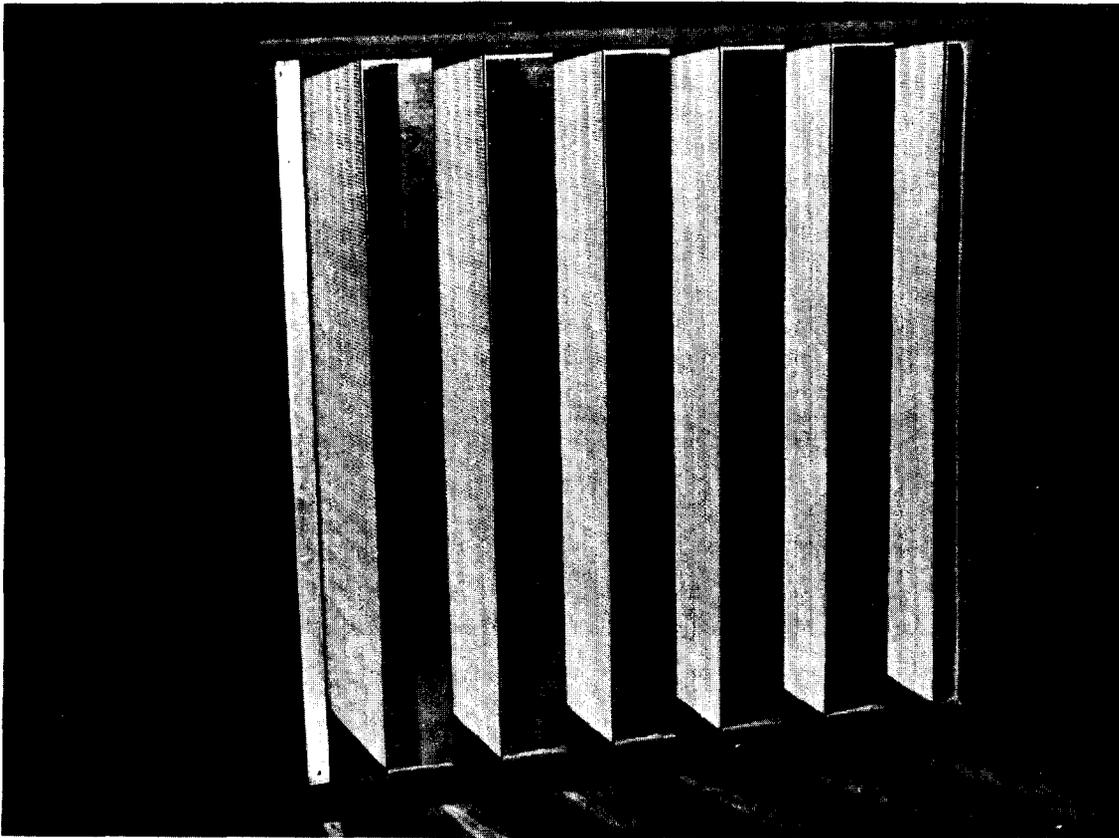


Figure 1. HEPA filter of continental European design.

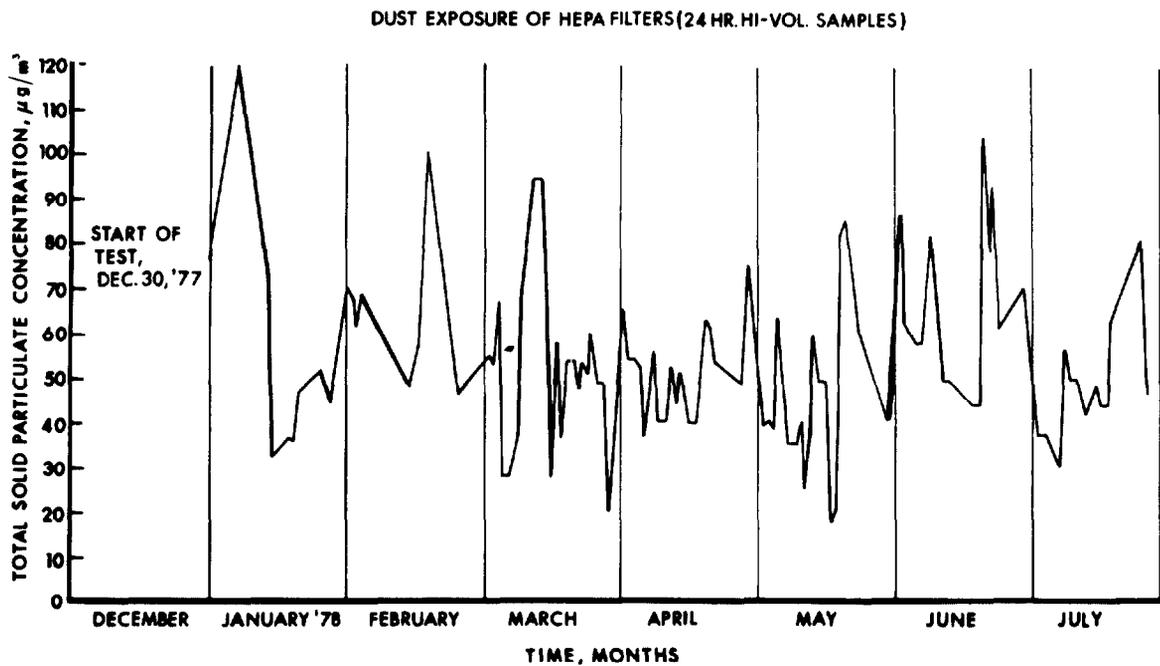


Figure 2. Dust exposure of HEPA filters (24 hr. Hi-Vol samples).

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size will be a close match for the aerosols these filters are likely to be exposed to in practice. Therefore, when a decision was made to test the life extension properties of European-design HEPA filters, a test location was selected on the open roof of the 15-story building that houses the Harvard Air Cleaning Laboratory. Dust concentrations at this site are measured daily with high volume air samplers using a technique published by the U.S. EPA and widely used throughout the world for 24-hour average total suspended particulated matter determinations. Figure 2 shows 24 hour average values since December 1977 at this location. Dust concentration fluctuates between 30 and 95 micrograms total dust per cubic meter of air ($\mu\text{g}/\text{m}^3$) with an average of $55 \mu\text{g}/\text{m}^3$ over a period of approximately 7 months.

The test is designed to compare the service life of an American-design filter operated at 1,000 cfm with a European-design filter operated at its rated flow rate of 1800 cfm and at a downrated flow-rate of 1000 cfm. For this purpose, three identical 24 x 24 x 11 5/8 in. filter test rigs have been constructed side by side on the roof, Figure 3. Each consists of (1) an inlet louvered grill to exclude snow and rain, (2) a straight 24 x 24 inch cross-section entry duct 4 feet long to straighten flow before entry to the filter and provide greater protection of the filter from the elements, (3) the test filter clamped between flanges with filter edge gaskets on both faces, (4) a flow regulating slide damper and, (5) an exhaust blower-motor set of appropriate size for the desired flow rate at resistance in excess of 4 in.w.g. across the test filter. Airflow resistance of the entry grill has been calibrated to measure airflow rate. The installation is shown in Figure 3.

European-design filters were purchased from Luwa Co. of Switzerland. They are designated "Luwa Standard Cells, Type N" and contain 35 m² of active glass paper filter surface. The filter paper used in these Luwa filters is manufactured by Dexter Corp., Windsor Locks, Connecticut and was not, then, used by any U.S. manufacturer of HEPA filters. Therefore, it was necessary to obtain a roll of this paper from Dexter Corp. and have it fabricated into U.S. filters corresponding to MIL-F-51068D. American Air Filter Co. of Louisville, KY fabricated our 1,000 cfm filters with aluminum separators, rubber base adhesive, and the Dexter/Luwa glass filter paper.

Results

The test filters were installed, tested in-place with DOP, and they have been in continuous operation since December 1977 at the pre-selected airflow rates. Filters are inspected daily, airflow readjusted, when needed, and filter pressure drop noted. At the same time, the 24-hour high volume air sampling filter is changed. High volume air sampling is maintained on a continuous basis over weekends and holidays. Figure 4 shows the starting (clean) resistance of each filter and the increase of resistance with time to the end of July 1978. It will be apparent from the figure that several reversible resistance increases have occurred over the test period; the most marked during January 1978. This peak resulted from a heavy snow fall that partially covered the louvered inlet grills and permitted snow to reach the filter inside, but after drying, airflow resistance returned to its former level. The later temporary

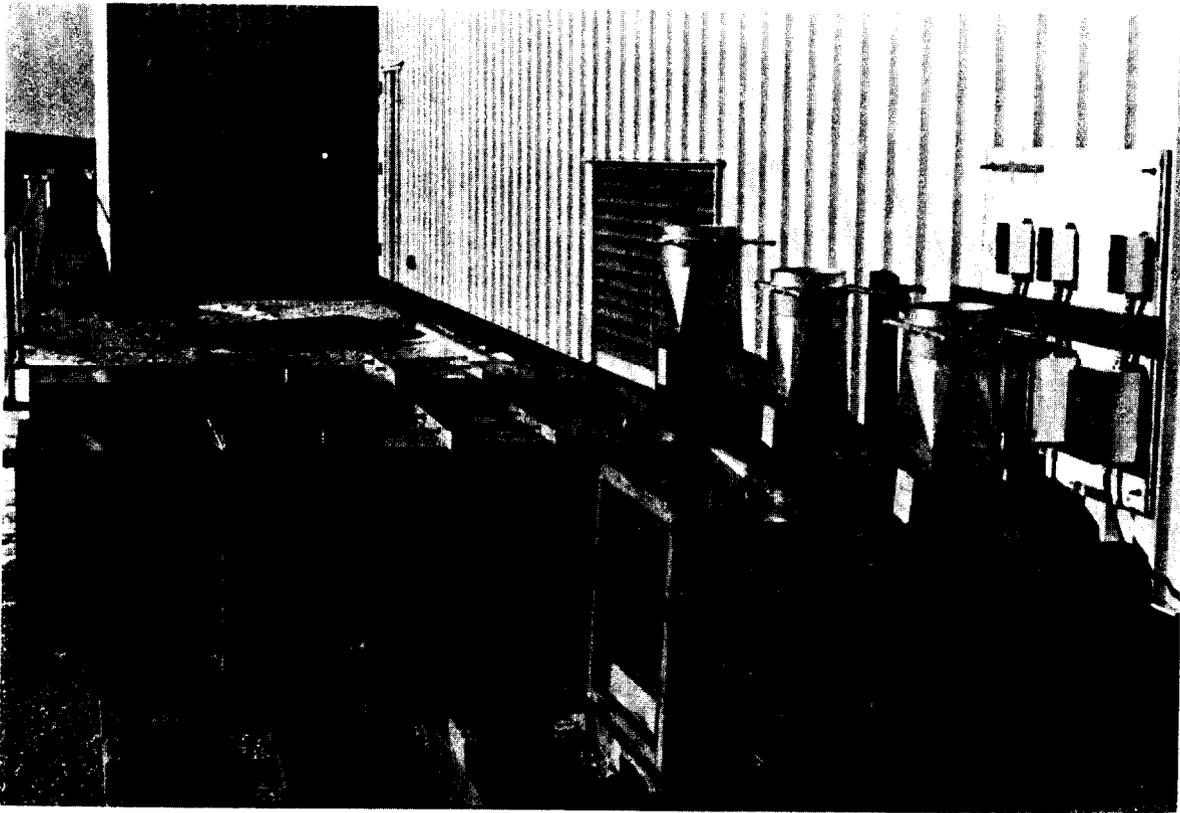


Figure 3. HEPA filter test rigs on roof on 15 story building.

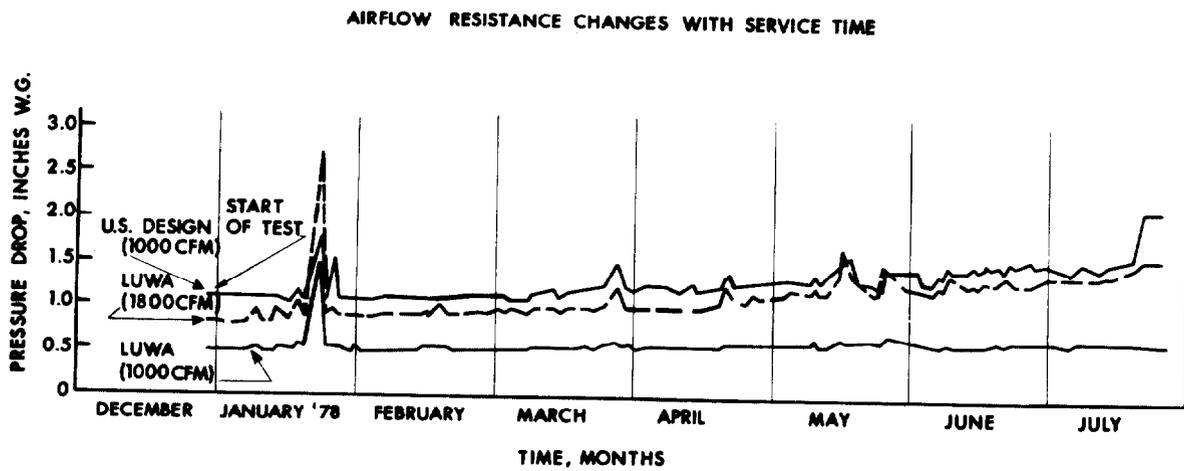


Figure 4. Airflow resistance changes with service time.

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peaks are related to periods of heavy rain.

A total of 459 grams (1 lb.) of atmospheric dust has entered each of the filters operated at 1,000 cfm and 826 grams (1.8 lb.) has entered the 1,800 cfm filter. One gram of dust has been recovered from the bottom of the entry duct leading to the 1,800 cfm filter and two grams from each of the entry ducts leading to the filters operated at 1,000 cfm. It is assumed that the remainder has deposited on the surfaces of the filters. The U.S.-design 1,000 cfm filter had an initial airflow resistance of 1.1 in. w.g. and this had increased to 2.2 in. w.g. by July 31, 1971. The 1,000 cfm Luwa filter (rated at 1800 cfm) started clean at 0.50 in. w.g. and has increased to 0.66 in. w.g. The Luwa filter operated at 1800 cfm started with a clean resistance of 0.83 in. w.g. and this has increased to 1.6 in. w.g. with the accumulation of 1.8 lb. of dust. From the figure, it will be clear that the U.S. filter and the 1800 cfm Luwa filter are responding in a parallel manner to the accumulation of dust but that the Luwa filter operating at 1,000 cfm is generating a resistance curve with a much flatter slope.

Future Plans

It is intended that this test will continue for the next two years and that the filters currently under test will be joined this month by three additional filters: one Sofiltra-Poelman unit of European design (Filtru A Air) operated at 1800 cfm and one operated at 1,000 cfm plus a new high capacity (2,000 cfm) filter manufactured by the American Air Filter Co. It is intended that these, also, will be operated for at least two years under the exposure conditions that have been described.

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DECONTAMINATION OF HEPA FILTERS

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Abstract

Mound Facility, during many years of plutonium-238 experience, has recovered over 150 kg of plutonium-238. Much of this material was recovered from HEPA filters or from solid wastes such as sludge and slag. The objective of this task was to modify and improve the existing nitric acid leaching process used at Mound so that filters from the nuclear fuel cycle could be decontaminated effectively. Various leaching agents were tested to determine their capability for dissolving PuO_2 , UO_2 , U_3O_8 , AmO_2 , NpO_2 , CmO_2 , and ThO_2 in mixtures of the following: HNO_3 -HF; HNO_3 -HF- H_2SO_4 ; and HNO_3 - $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$. Adsorption isotherms were obtained for two leaching systems. In some tests simulated contaminated HEPA filter material was used, while in others actual spent glovebox filters were used.

The maximum decontamination factor of 833 was achieved in the recovery of plutonium-238 from actual filters. The dissolution was accomplished by using a six-stage process with 4N HNO_3 -0.23M $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$ as the leaching agent. Thorium oxide was also effectively dissolved from filter media using a mixture of nitric acid and ceric ammonium nitrate. Sodium carbonate and Na_2CO_3 - KNO_3 fusion tests were performed using simulated PuO_2 -contaminated filter media at various temperatures. Approximately 70 wt% of the PuO_2 was soluble in a mixture composed of 70 wt% Na_2CO_3 -30 wt% KNO_3 (heated for 1 hr at 950°C).

Introduction

Efforts were directed toward determining the dissolution parameters of PuO_2 , UO_2 , and U_3O_8 ** in various leaching reagents. The reagents used were various concentrations of HNO_3 , HNO_3 -HF, HNO_3 -HF- H_2SO_4 , HNO_3 - $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$, and HNO_3 - H_2SO_4 . Two types of plutonium dissolution tests were conducted with 8N HNO_3 . In the first test, PuO_2 was placed in 8N HNO_3 at boiling temperature. In the other test, HEPA filter media contaminated with PuO_2 were added to boiling 8N HNO_3 . The dissolution rates of PuO_2 as measured by the alpha activity of the acid media were determined for each test; as shown in Figure 1, the apparent dissolution rate was less in the test using PuO_2 -contaminated filter media. After 14 hr of heating, for example, there was a 3% difference (11% minus 8%) in dissolution rates. At this point, plutonium adsorption isotherms were developed to determine whether any of the dissolved plutonium was being adsorbed by the filter media.

*Mound Facility is operated by Monsanto Research Corporation for the U.S. Department of Energy under Contract No. EY-76-C-04-0053.

**The uranium oxides were depleted uranium-238, whereas the PuO_2 was a mixture of 80 wt% plutonium-238, 16 wt% plutonium-239, 2.5 wt% plutonium-240, 0.8 wt% plutonium-241, 0.2 wt% plutonium-242, plus small amounts of other actinides calcined at 750°C.

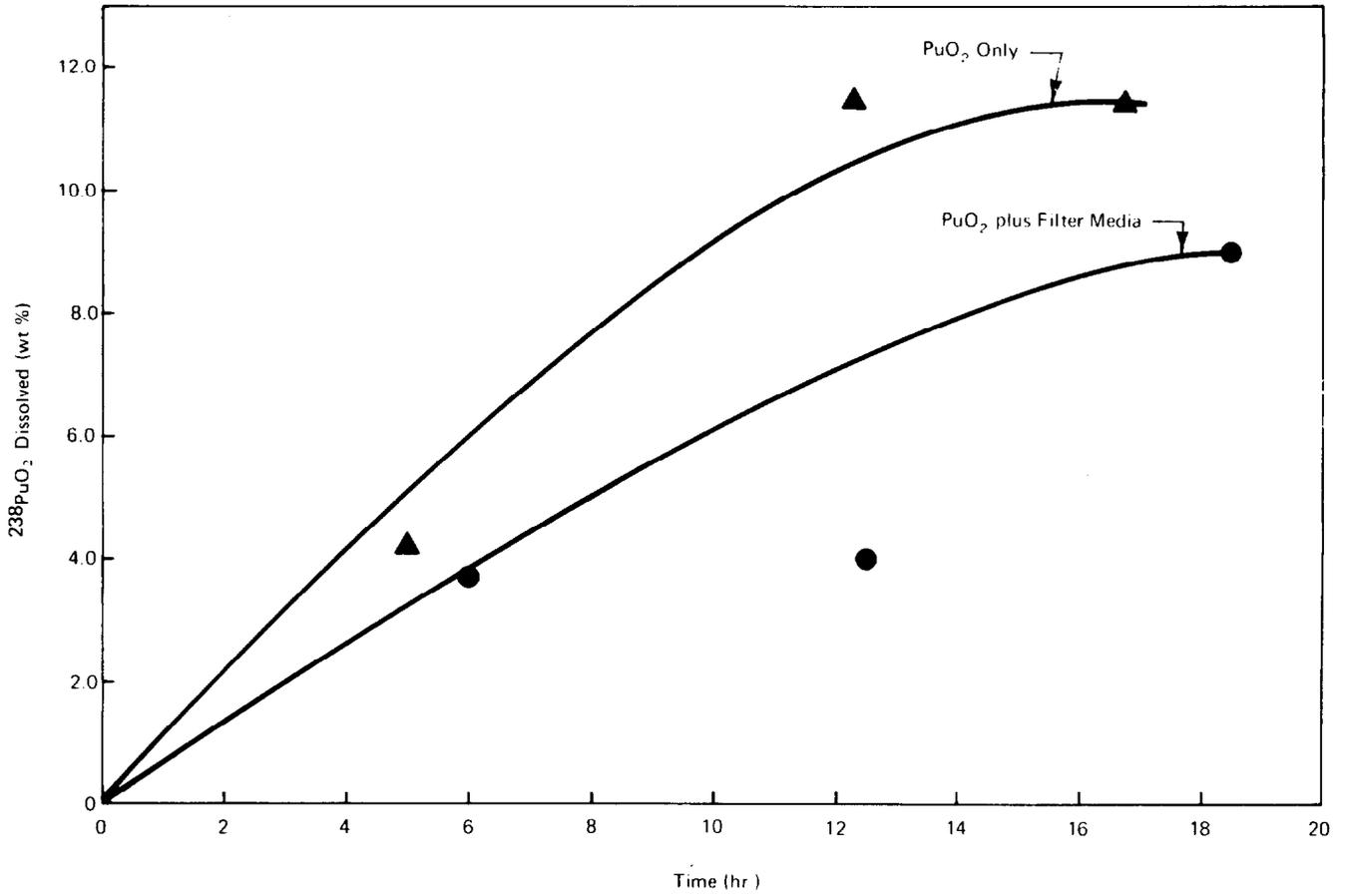


Figure 1 - Effect of filter media on dissolution rates of PuO_2 in boiling 8N HNO_3 .

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This is of great importance since the amount of adsorbed plutonium would limit the decontamination factor that could be achieved. Adsorption isotherms were determined for the Pu-HNO₃ and Pu-HNO₃-HF systems.

Efforts were also directed toward the determination of dissolution parameters in various reagents of PuO₂* and PuO₂-UO₂** solid solution. The reagents used were various concentrations of HNO₃, -HF-H₂SO₄, HNO₃-HF, HNO₃-(NH₄)₂Ce(NO₃)₆, and HNO₃-H₂SO₄. In one series of tests, simulated contaminated HEPA filter media was used. This material was prepared by mixing shredded filter media with actinide oxides. In another series, actual HEPA glovebox filter media contaminated with PuO₂ were used. Fusion tests were also completed using simulated PuO₂-contaminated filter media. Both Na₂CO₃ and Na₂CO₃-KNO₃ were investigated as possible fusion agents.

Dissolution parameters (in various reagents) of americium-241 and plutonium-239 oxide mixtures, uranium-233 oxide, neptunium-237 oxide, curium-244 oxide, thorium-232 oxide, and PuO₂ were determined. The reagents used were various concentrations of HNO₃-HF-H₂SO₄, HNO₃-(NH₄)₂Ce(NO₃)₆, HNO₃-HF and HNO₃. Both simulated contaminated HEPA filter media and actual glovebox filter media from spent filters were used. The maximum decontamination factor was 833, achieved using a six-stage dissolution process. Also, PuO₂ was fused with Na₂CO₃ various elevated temperatures, and a dissolution percentage was determined.

Experimental Work

Adsorption Studies

Standard plutonium solutions were prepared by dissolving PuO₂ in boiling 8N HNO₃. The HEPA filter media were shredded and ball-milled to <40 mesh. The filter media were placed in 100 ml of the prepared PuO₂-acid solution and stirred magnetically for several hours to reach equilibrium. A blank sample was run without filter media to correct for adsorption on the test beakers. All tests were made at ambient temperature (≈20°C).

Figure 2 shows the adsorption of plutonium on filter media in different acids. The ordinate is x/m, where x = milligrams of plutonium-238 adsorbed and m = milligrams of filter media. The abscissa is C, which is the concentration of the solution in equilibrium with the

*The uranium oxides were depleted uranium-238, whereas the PuO₂ was a mixture of 80 wt% Plutonium-238, 16 wt% Plutonium-239, 2.5 wt% Plutonium-240, 0.8 wt% Plutonium-241, 0.2 wt% Plutonium-242, plus small amounts of other actinides calcined at 750°C.

**The uranium was uranium-238 while the plutonium was 85.0 wt% plutonium-239, 12.0 wt% plutonium-240, 1.7 wt% plutonium-241, 0.75 wt% plutonium-238, plus small amounts of other actinides.

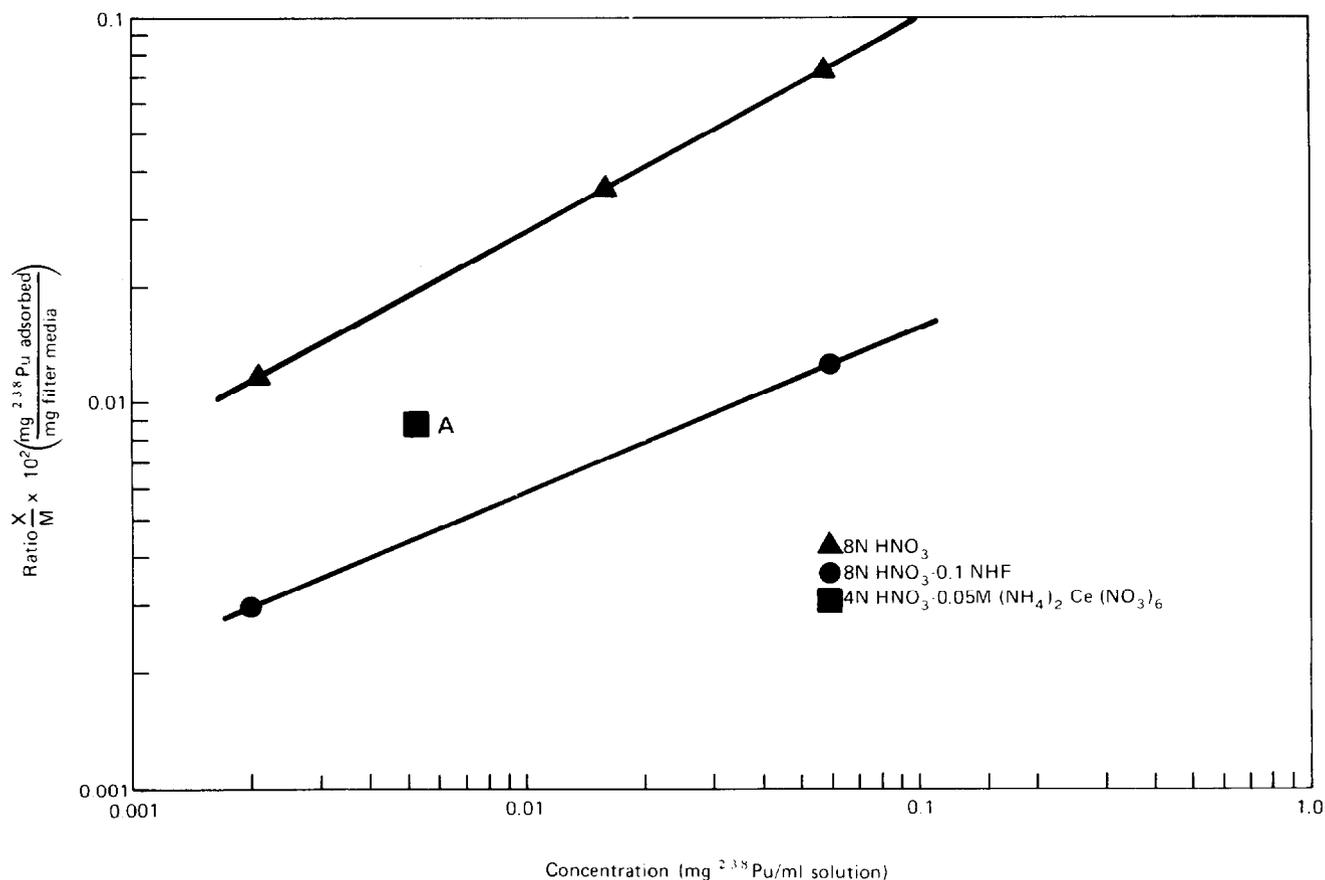


Figure 2 - Adsorption of ²³⁸Pu on filter media at ambient temperature (~20°C) in different acids.

filter media (expressed in milligrams of plutonium-238 per milliliter of solution). The Freundlich equation relates x/m and C as follows:

$$x/m = kC^{1/n} \quad (1)$$

or

$$\log (x/m) = \log k + (1/n) \log C \quad (2)$$

The plot of $\log (x/m)$ as a function of $\log C$ should be a straight line with slope $1/n$ and $\log k$ the point of intercept. The straight lines in Figure 2 are such a plot. It should be noted that for any given concentration, the x/m ratio is smaller for the 8N HNO₃-0.1N HF system. This means that a larger decontamination factor could be achieved using the HNO₃-HF leaching agent (assuming that the same percentage of PuO₂ is dissolved by the reagent in the dissolution step).

Tests were also performed to determine the solubility of the filter media in various dissolution reagents. It was determined that approximately 30 wt% of the filter media is soluble in 8N HNO₃ and approximately 50 wt% of the filter media dissolves in 8N HNO₃-0.1N HF. Also, in another adsorption test it was found that the use of 4N HNO₃-0.05M ceric ammonium nitrate (see point A, Figure 2) would result in a decontamination factor between that obtained with 8N HNO₃-0.1N HF and that with 8N HNO₃ (assuming each leaching agent was equally effective in dissolving PuO₂).

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PuO₂ Dissolution Studies

Contaminated HEPA filter media were prepared by mixing PuO₂* powder with shredded filter media. The plutonium-238 concentration of the mixture was 11 mg/cm³ of prepared media. Small samples of this prepared mixture (about 3 g) were added to glass beakers containing 250 ml of leaching solution. All tests were performed at boiling temperatures. Samples were withdrawn periodically and the plutonium-238 concentration of the solution was determined. The contents of the beakers were stirred every 2 hr. The solution volume and concentration were kept constant by adding acid of proper concentration in order to replace evaporated acid.

Table I lists results from dissolution tests for PuO₂ in various acid mixtures. The HNO₃-H₂SO₄ mixtures were relatively ineffective dissolution agents as illustrated in Figure 3. A fivefold increase in H₂SO₄ concentration increased the dissolution rate by only 3%. The maximum weight percent of plutonium-238 dioxide dissolved was only 18% after an 18-hr reaction time. Figure 4 illustrates the effect of initial cerium(IV) concentration on the dissolution rate. The 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆ leaching agent was by far the most effective of the three HNO₃-cerium(IV) solutions, with 88% of the plutonium-238 dioxide dissolved in 3 hr.

The reaction mechanism for dissolving PuO₂ in HNO₃-cerium(IV) solutions is shown in Equations 3 and 4.



Because stoichiometric quantities of cerium(IV) are required for complete dissolution of the PuO₂, large amounts of cerium(IV) are required to dissolve large quantities of PuO₂. Therefore, it would seem advantageous to use small amounts of cerium(IV) and then add oxidizing compounds to oxidize the cerium(III) to cerium(IV). An example of this would be the addition of KMnO₄ to the depleted solution. This would cause the Mn⁺⁷ + 3e⁻ ⇌ Mn⁺⁴ reaction which would oxidize the depleted cerium as follows:



where one mole of manganese oxidizes three moles of cerium. This was done in acid tests 25 and 25A (see Table I). Approximately 1 g of KMnO₄ was added to a depleted 4N HNO₃-0.005M(NH₄)₂Ce(NO₃)₆ solution and the solution heated an additional 15 hr. In this period of time, the concentration of PuO₂ dissolved increased significantly from 12 wt% in the depleted solution to 26 wt% in the regenerated solution.

*The PuO₂ was fine powder having a composition of 80 wt% plutonium-238, 16 wt% plutonium-239, 2.5 wt% plutonium-240, 0.8 wt% plutonium-241, 0.2 wt% plutonium-242, plus small amounts of other actinides. It was calcined for 2 hr at 950°C.

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Table I - DISSOLUTION OF PLUTONIUM DIOXIDE IN VARIOUS ACID MIXTURES

Acid Test Number	Reagent Composition ^a	Time Heated (hr)	PuO ₂ Dissolved (wt %)
19-1	4N HNO ₃ -0.05M CAN	5 3/4	42.9
19-2	4N HNO ₃ -0.05M CAN	12 1/4	41.4
19-3	4N HNO ₃ -0.05M CAN	18 3/4	38.5
20-1	8N HNO ₃ -0.1N H ₂ SO ₄	5 3/4	14.4
20-2	8N HNO ₃ -0.1N H ₂ SO ₄	12	12.7
20-3	8N HNO ₃ -0.1N H ₂ SO ₄	18	15.9
21-1	12N HNO ₃ -0.1N HF-0.1N H ₂ SO ₄	4 3/4	115.2
21-2	12N HNO ₃ -0.1N HF-0.1N H ₂ SO ₄	10 1/2	107.1
22-1	12N HNO ₃ -0.1N HF	1 3/4	87.0
22-2	12N HNO ₃ -0.1N HF	6 3/4	94.1
22-3	12N HNO ₃ -0.1N HF	11 1/2	107.9
25-1	4N HNO ₃ -0.005M CAN	2 1/4	11.1
25-2	4N HNO ₃ -0.005M CAN	7 3/4	13.4
25-3	4N HNO ₃ -0.005M CAN	13 1/4	11.9
25-A-1	4N HNO ₃ -0.005M CAN- KMnO ₄	3	13.4
25-A-2	4N HNO ₃ -0.005M CAN- KMnO ₄	8 1/2	18.9
25-A-3	4N HNO ₃ -0.005M CAN- KMnO ₄	14 3/4	26.3
26-1	12N HNO ₃ -0.01N HF	1 3/4	31.6
26-2	12N HNO ₃ -0.01N HF	7 1/4	41.0
26-3	12N HNO ₃ -0.01N HF	12 3/4	40.9
27-1	12N HNO ₃ -0.01N HF-0.1N H ₂ SO ₄	1 1/4	30.1
27-2	12N HNO ₃ -0.01N HF-0.1N H ₂ SO ₄	6 3/4	33.1
27-3	12N HNO ₃ -0.01N HF-0.1N H ₂ SO ₄	12 1/4	36.3
28-1	12N HNO ₃ -0.1N HF-0.01N H ₂ SO ₄	1	96.3
28-2	12N HNO ₃ -0.1N HF-0.01N H ₂ SO ₄	6 1/2	100.9
28-3	12N HNO ₃ -0.1N HF-0.01N H ₂ SO ₄	12	101.8
29-1	8N HNO ₃ -0.5N H ₂ SO ₄	5 1/2	15.8
29-2	8N HNO ₃ -0.5N H ₂ SO ₄	10 3/4	17.4
29-3	8N HNO ₃ -0.5N H ₂ SO ₄	15 1/4	17.4
30-1	12N HNO ₃ -0.05N HF 0.01N H ₂ SO ₄	3	95.9
30-2	12N HNO ₃ -0.05N HF 0.01N H ₂ SO ₄	8 1/4	88.0
30-3	12N HNO ₃ -0.05N HF 0.01N H ₂ SO ₄	14 1/2	97.7
31-1	4N HNO ₃ -0.1M CAN	3	87.7
31-2	4N HNO ₃ -0.1M CAN	8 1/4	88.0
31-3	4N HNO ₃ -0.1M CAN	14 1/4	80.6
32-1	8N HNO ₃ -K ₂ S ₂ O ₈	3	0.86
32-2	8N HNO ₃ -K ₂ S ₂ O ₈	7 1/2	0.82
32-3	8N HNO ₃ -K ₂ S ₂ O ₈	13 1/2	1.89

^aCAN is abbreviation for ceric ammonium nitrate

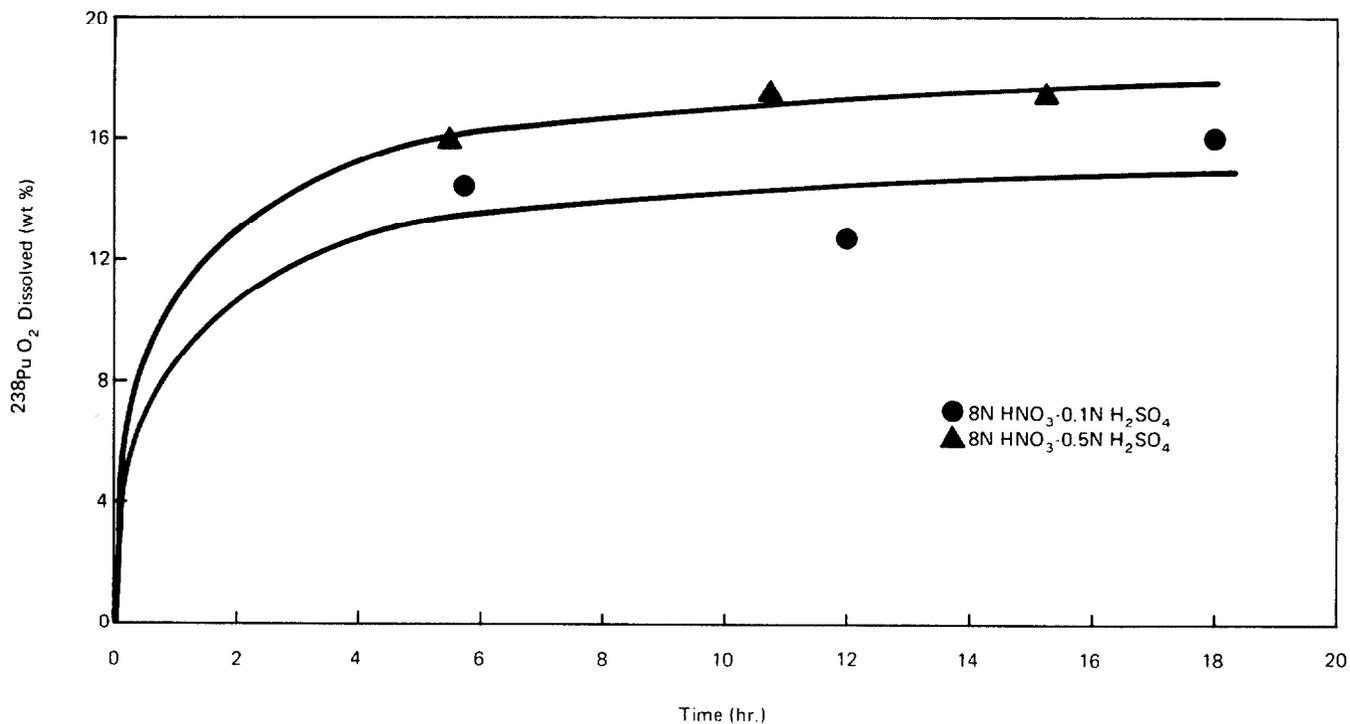


Figure 3 - Effect of H₂SO₄ concentration on dissolution rate.

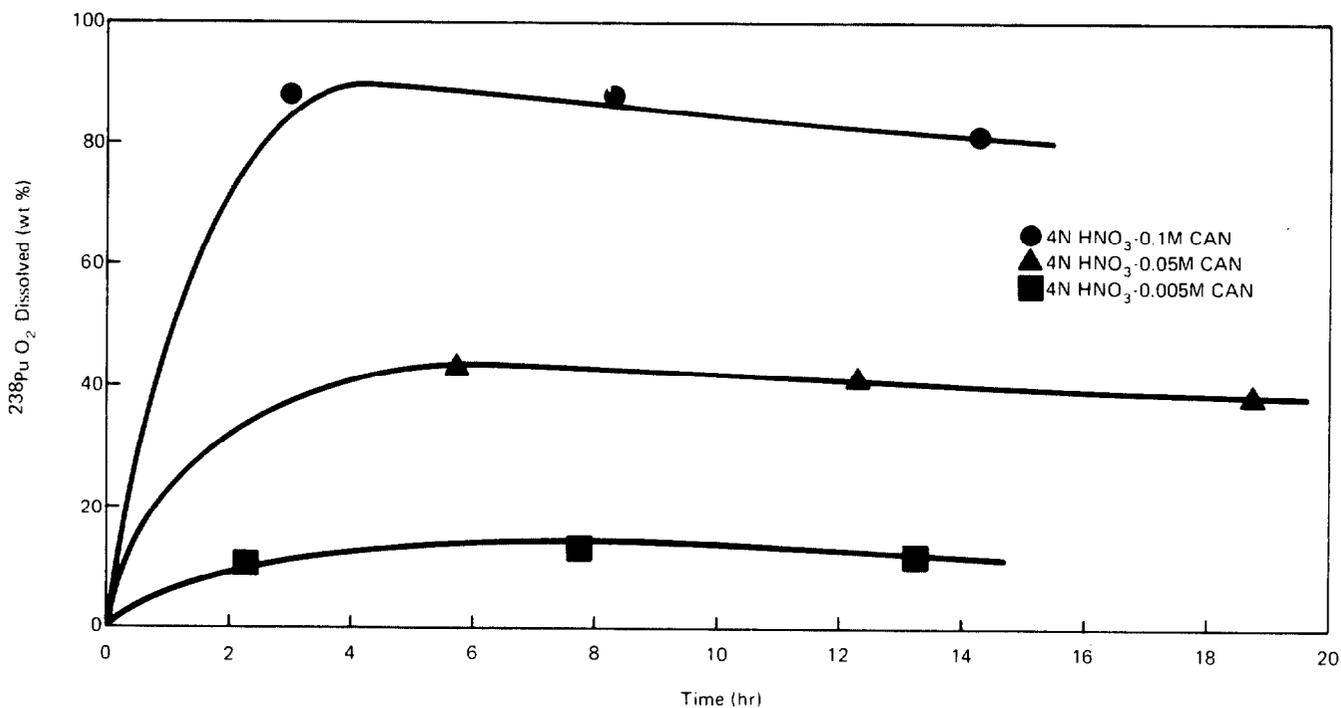


Figure 4 - Effect of ceric ammonium nitrate (CAN) concentration on dissolution rate.

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This method does, however, add manganese and potassium to the dissolved salts of the waste stream.

Another experiment was conducted using 8N HNO_3 and $\text{K}_2\text{S}_2\text{O}_8$ (potassium persulphate) as an oxidizing agent (see Table I, acid test 32). After 13½ hr of reaction time at boiling temperature, only 1.89% of the PuO_2 had dissolved. Figure 1 shows that approximately 7.5% of the PuO_2 dissolved in 13½ hr with no additional oxidizing agent present. Thus the $\text{K}_2\text{S}_2\text{O}_8$ actually decreased the dissolution rate of the PuO_2 .

Figures 5 and 6 show the effect of the HF concentration on the dissolution rate of PuO_2 . For the HNO_3 -HF system, a tenfold increase in the HF concentration increases the concentration of dissolved PuO_2 from about 40 wt% to approximately 100 wt% for 9 hr of reaction time (Figure 5). A twofold increase in the HF concentration for the HNO_3 -HF- H_2SO_4 system increases the concentration of dissolved PuO_2 from 94 wt% to 100 wt% for 9 hr of reaction time (Figure 6). As can be seen in Table I, any dissolution test in which the HF concentration was 0.05N, or greater, was successful in obtaining a 90 to 100% dissolution of the PuO_2 . The effect of sintering temperature on dissolution rate for various reagents can be seen in Figures 7 and 8.

UO_2 and U_3O_8 Dissolution Studies

Contaminated filter media samples were prepared by mixing 0.2 g of either UO_2 or U_3O_8 * with 2.8 g of shredded filter media. These samples were combined in a glass round-bottom flask containing 250 ml of the desired leaching solution. The flasks were attached to reflux condensers, and the contents were heated and refluxed at boiling temperatures. Samples were withdrawn periodically, and the uranium concentration of the solution was determined.

Table 2 lists results of dissolution tests for uranium oxide in various acid mixtures; in every test the uranium oxides dissolved rapidly. The percentage of uranium oxide dissolved was always greater than 90%; 8N HNO_3 performed as well as any of the acid mixtures, dissolving essentially 100% of the UO_2 or U_3O_8 after 6 hr of heating. As a comparison, the percentage of PuO_2 dissolved after 6 hr was ~4% (see Figure 1).

PuO_2 -75 wt% UO_2 Solid Solution Studies

The solid solution was composed of plutonium** and uranium-238 oxides which had been fired at 1600°C in a reducing atmosphere. Six dissolution tests were performed (see Table III). The powdered PuO_2 - UO_2 solid solution was mixed thoroughly with shredded filter media using a ratio of 1 g of solid solution to 12 g of filter media. A small sample of the mixture (approximately 3 g) was added to a glass beaker containing 250 ml of leaching solution. Samples were withdrawn periodically

*The U_3O_8 had been fired at 950°C for 2 hr.

**Assay of the plutonium was 85.0 wt% plutonium-239, 12.0% plutonium-240, 1.7% plutonium-241, 0.75% plutonium-238, plus small amounts of other actinides.

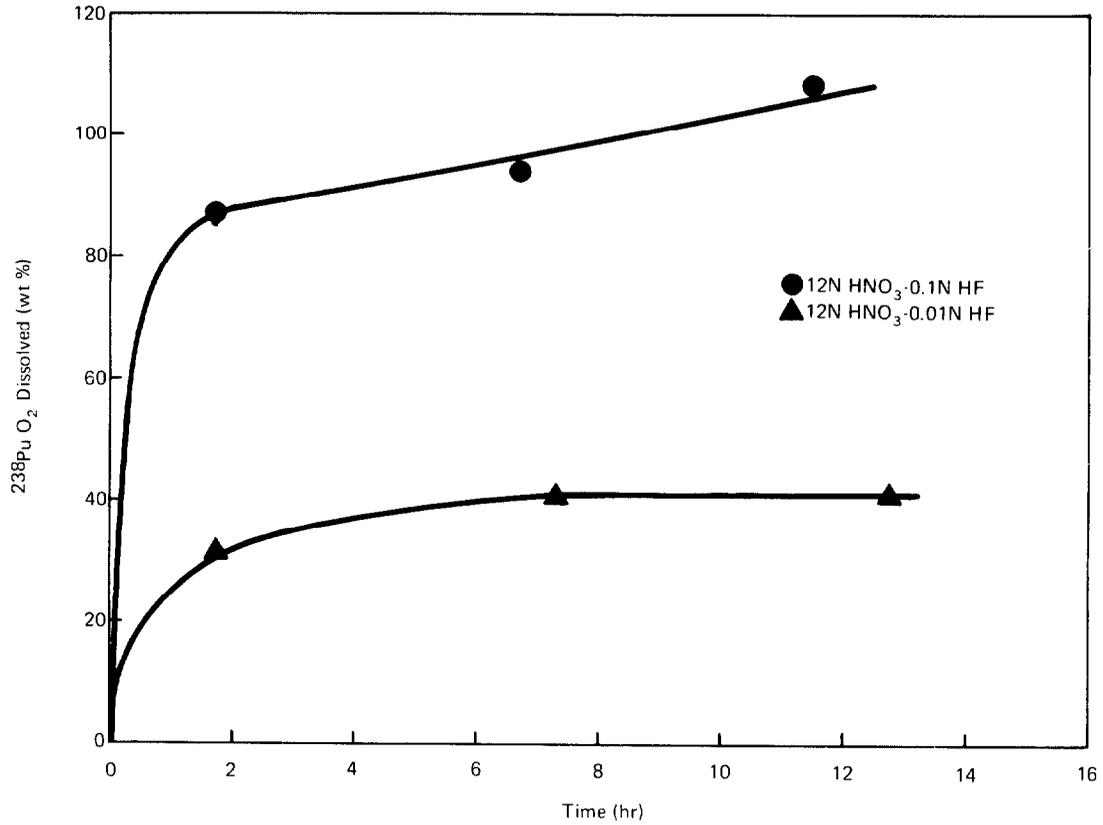


Figure 5 - Effect of HF concentration on dissolution rate of PuO₂.

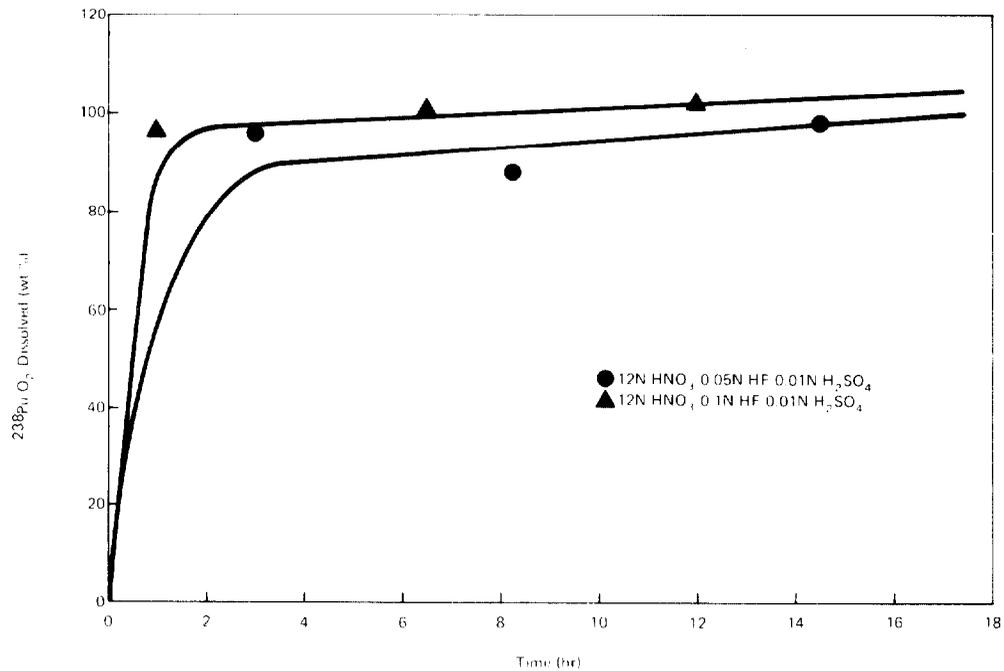


Figure 6 - Effects of HF and H₂SO₄ concentrations on dissolution rates.

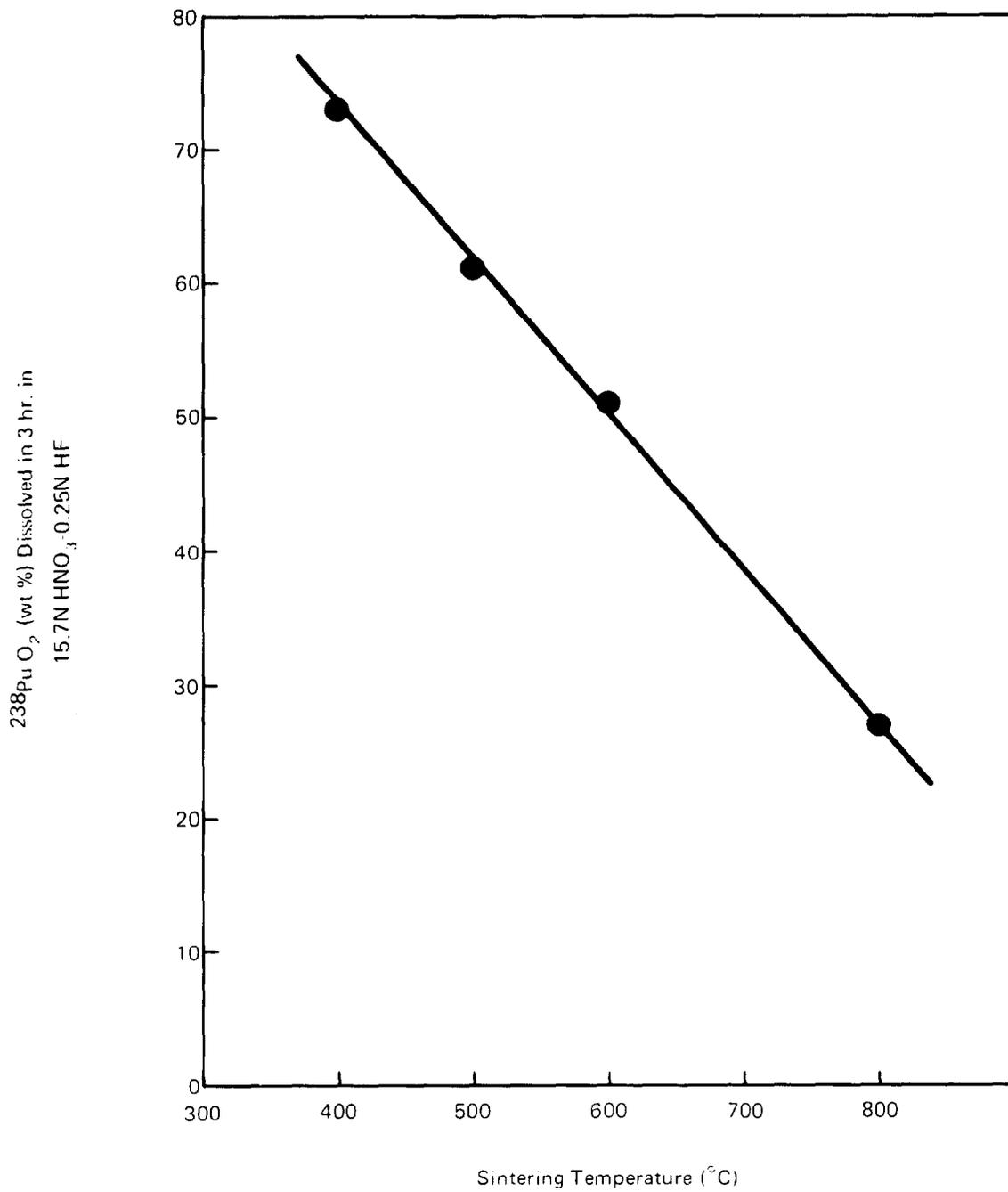


Figure 7 - Effect of sintering temperature on dissolution rate (2).

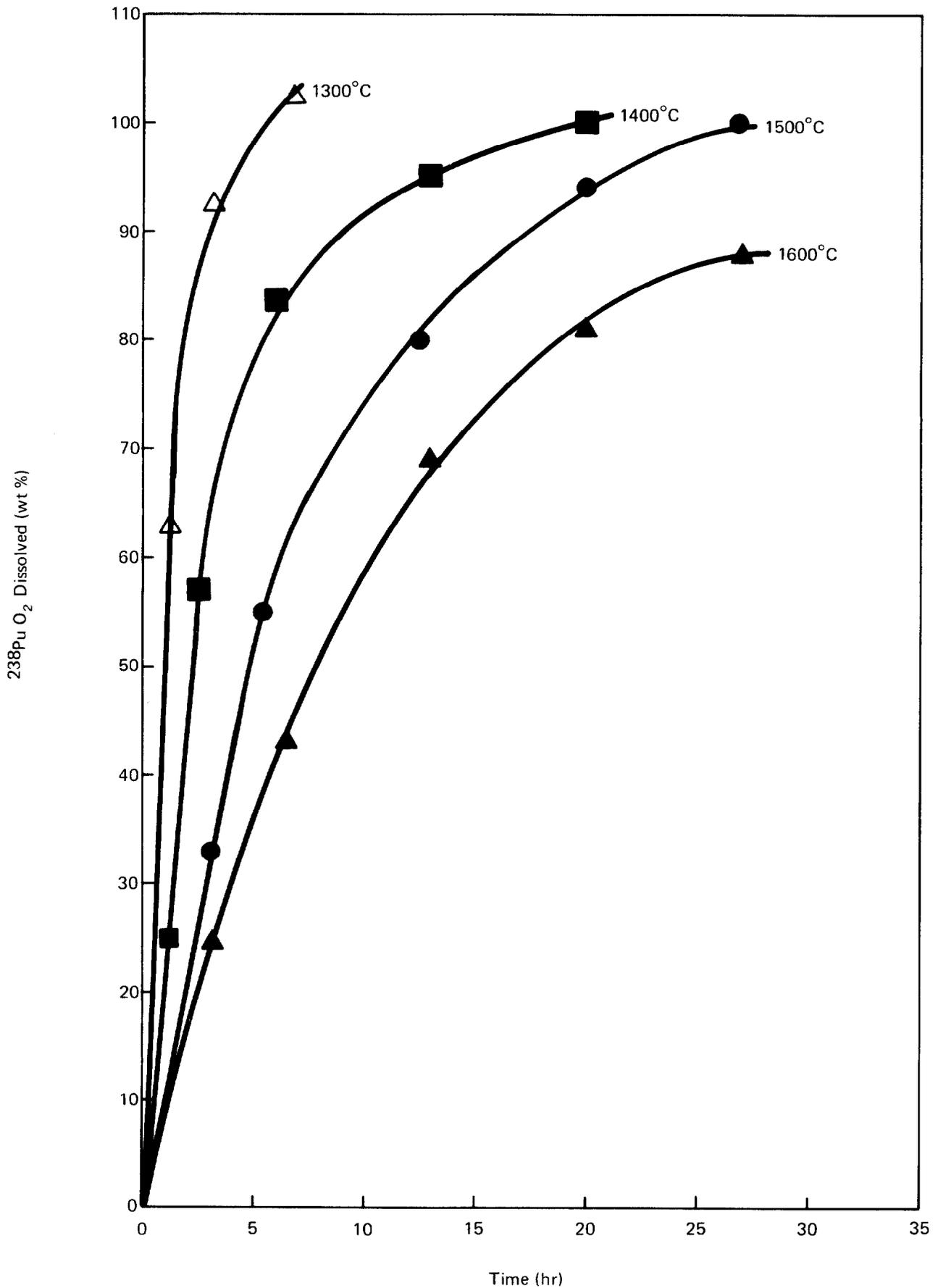


Figure 8 - Effect of sintering temperature on plutonium dioxide dissolution in 4N HNO_3 - 0.15M cerium(IV) (3).

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Table II - DISSOLUTION OF URANIUM OXIDES IN VARIOUS ACID MIXTURES

Acid Test Number	Reagent Composition	Time Heated (hr)	UO ₂ or U ₃ O ₈ Dissolved (wt %)
UO ₂ -1-1	12N HNO ₃ -0.01N HF-0.1N H ₂ SO ₄	1/2	92.8
UO ₂ -1-2	12N HNO ₃ -0.01N HF-0.1N H ₂ SO ₄	6 1/2	94.1
UO ₂ -2-1	12N HNO ₃ -0.1N HF-0.01N H ₂ SO ₄	1	91.0
UO ₂ -2-2	12N HNO ₃ -0.1N HF-0.01N H ₂ SO ₄	7	99.6
UO ₂ -3-1	12N HNO ₃ -0.1N H ₂ SO ₄	4 3/4	99.3
UO ₂ -4-1	8N HNO ₃	5 3/4	102.8
U ₃ O ₈ -5-1	8N HNO ₃	1	107.0
U ₃ O ₈ -6-1	12N HNO ₃ -0.1N H ₂ SO ₄	1	103.0

and the percentage of solid solution dissolved was determined. The solution volume and concentration were kept constant by adding acid of proper concentration in order to replace evaporated acid.

As can be seen in Table III, four reagents were successful in dissolving greater than 93% of the PuO₂-UO₂ in 2 hr. These were 12N HNO₃-0.1N HF, 12N HNO₃-0.05N HF-0.01N H₂SO₄, 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆, and 8N HNO₃. Both the 4N HNO₃ and the 4N HNO₃-0.1N H₂SO₄ were unsuccessful in attaining a 90% dissolution even when leaching times of 7 hr were used. Figure 9 shows the effect of HNO₃ concentration on the dissolution rate. Doubling the acid normality (from 4N to 8N) doubled the percent dissolved (after 2 hr of heating) from 46% to 93%. Therefore, it can be concluded that 8N HNO₃, 12N HNO₃-0.1N HF, 12N HNO₃-0.05N HF-0.01N H₂SO₄, and 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆ would be acceptable leaching reagents for PuO₂-UO₂ solid solution whereas 4N HNO₃ and 4N HNO₃-0.1N H₂SO₄ are unacceptable.

Salt Fusion Studies

Sodium carbonate and Na₂CO₃-KNO₃ fusions were completed using PuO₂* contaminated filter media. Small samples were prepared by thoroughly mixing 0.1 g PuO₂ and 1.5 g of filter media. Approximately 13 g of salt (Na₂CO₃ or Na₂CO₃-KNO₃) was placed in a platinum crucible and 1.6 g of contaminated filter media was then added. This was mixed thoroughly and then heated slowly to 950°C. The crucible and contents were maintained at 950°C for 1 hr and then allowed to cool slowly to ambient temperature. The cooled melt was then removed from the crucible and dissolved in 4N HNO₃. The acid was maintained at boiling temperature for at least 1 hr and subsequently sampled for plutonium-238 concentration. Table IV tabulates the results achieved in these

*The PuO₂ was a fine powder, having a composition of 80 wt% plutonium-238, 16% plutonium-239, 2.5% plutonium-240, 0.8% plutonium-241, 0.2% plutonium-242, plus small amounts of other actinides, calcined at 950°C.

Table III - PuO₂-UO₂ Solid Solution Dissolution

Acid Test Number	Reagent Composition	Time Heated (hr)	PuO ₂ -UO ₂ Dissolved (wt %)
Pu-U-1	12N HNO ₃ -0.1N HF	2	100.0
Pu-U-2	12N HNO ₃ -0.05N HF-0.01N H ₂ SO ₄	2	97.6
Pu-U-3	4N HNO ₃ -0.1M CAN ^a	2	94.8
Pu-U-4A	8N HNO ₃	1	71.8
Pu-U-4B	8N HNO ₃	2	93.2
Pu-U-5A	4N HNO ₃	2-1/2	49.6
Pu-U-5B	4N HNO ₃	4-1/2	54.2
Pu-U-5C	4N HNO ₃	7	60.0
Pu-U-6	4N HNO ₃ -0.1N H ₂ SO ₄	3-3/4	80.2

^aAbbreviation for ceric ammonium nitrate.

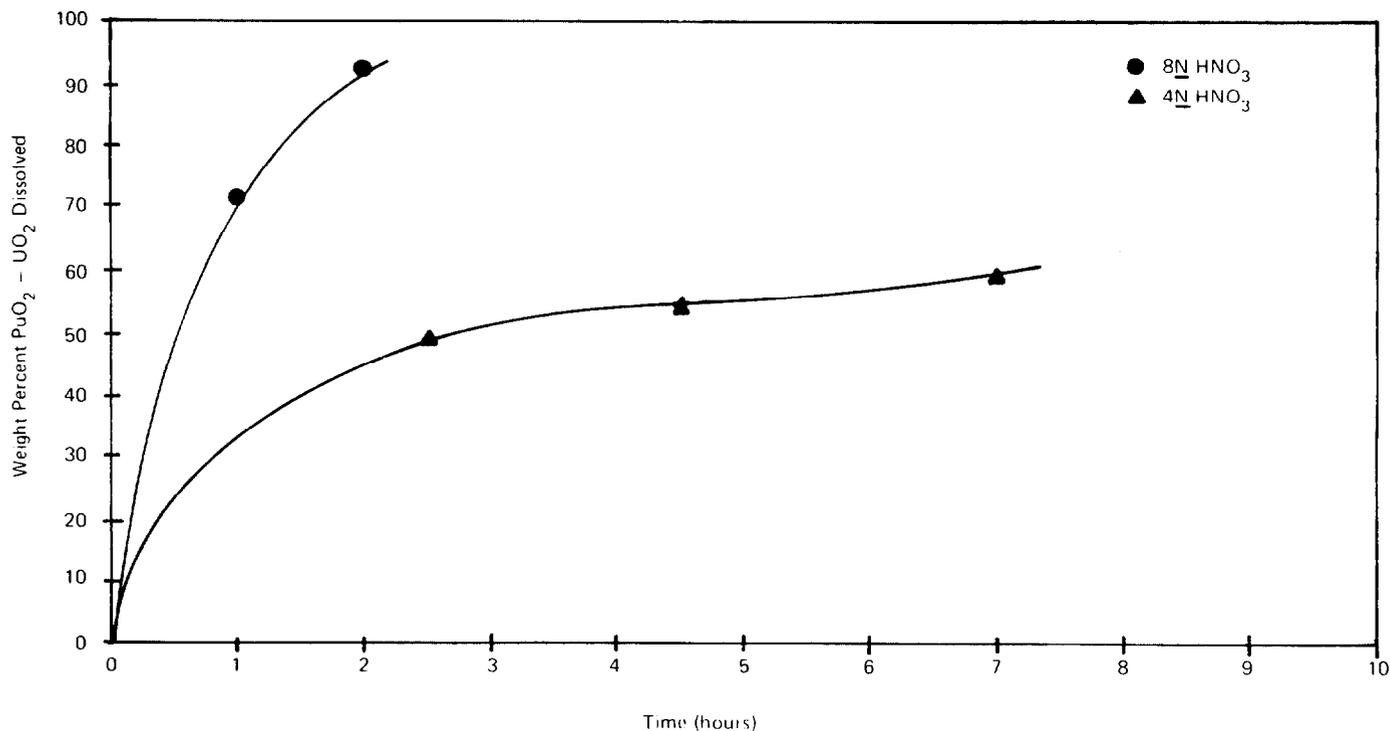


Figure 9 - Effect of HNO₃ concentration on dissolution rate of PuO₂.

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Table IV - Na₂CO₃-KNO₃ Fusion of PuO₂

Fusion Test Number	Salt Composition (wt %)	Time Heated (hr)	Operating Temperature (°C)	PuO ₂ Solubilized (wt %)
Pu-38	100% Na ₂ CO ₃	1	950	8.83
Pu-39	Na ₂ CO ₃ -8% KNO ₃	1	950	27.8
Pu-40	Na ₂ CO ₃ -16% KNO ₃	1	950	48.7
Pu-41	Na ₂ CO ₃ -30% KNO ₃	1	950	71.0

fusion experiments. As can be seen, the maximum percent of dissolution obtained was 71% using a salt mixture of Na₂CO₃-30 wt% KNO₃.

Figure 10 shows the effect of potassium nitrate composition on PuO₂ solubility. The effect is linear between zero and 16% KNO₃ and nonlinear at higher potassium nitrate concentrations. Also because of time limitations, only three Na₂CO₃-KNO₃ fusions were performed and the maximum concentration of potassium nitrate used was 30 wt%. It should be noted that the greater the concentration (wt%) of potassium nitrate, the more vigorous the reaction.

The temperature must be raised slowly in order to prevent the contents of the crucible from spilling over. The 71% recovery obtained using Na₂CO₃-30 wt% KNO₃ is considered unsatisfactory for our purposes. The dissolution must be greater than 90% in order to achieve large decontamination factors. Perhaps by using greater than 30% KNO₃ this result can be achieved.

Dissolution of Actual HEPA Glovebox Filter Media

Small test samples of approximately 3 g each were removed from an actual glovebox filter contaminated with PuO₂*. Each sample contained approximately 55 mg of plutonium-238. These samples were placed in a glass beaker containing 100 ml of leaching reagent. The leaching agents used were 4N HNO₃-0.1M ceric ammonium nitrate and 12N HNO₃-0.05N HF-0.01N H₂SO₄. The tests were performed at boiling temperatures. The contents of the beakers were stirred every hour, and samples were withdrawn periodically for plutonium analysis. The solution volume and acid concentration were kept constant by adding acid of proper concentration in order to replace evaporated acid. Table V contains the results obtained in these experiments. The average PuO₂ dissolution was 92.5 wt% using 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆ and 93.3 wt% using 12N HNO₃-0.05N HF-0.01N H₂SO₄. These compare well with the average dissolution percentages found for simulated filter media samples listed in Table I (85.4% and 93.9% respectively). Figure 11 is a graph of the dissolution rate for PuO₂ in

*The PuO₂ was a fine powder, having a composition of 80 wt% plutonium-238, 16% plutonium-239, 2.5% plutonium-240, 0.8% plutonium-241, 0.2% plutonium-242, plus small amounts of other actinides, calcined at 950°C.

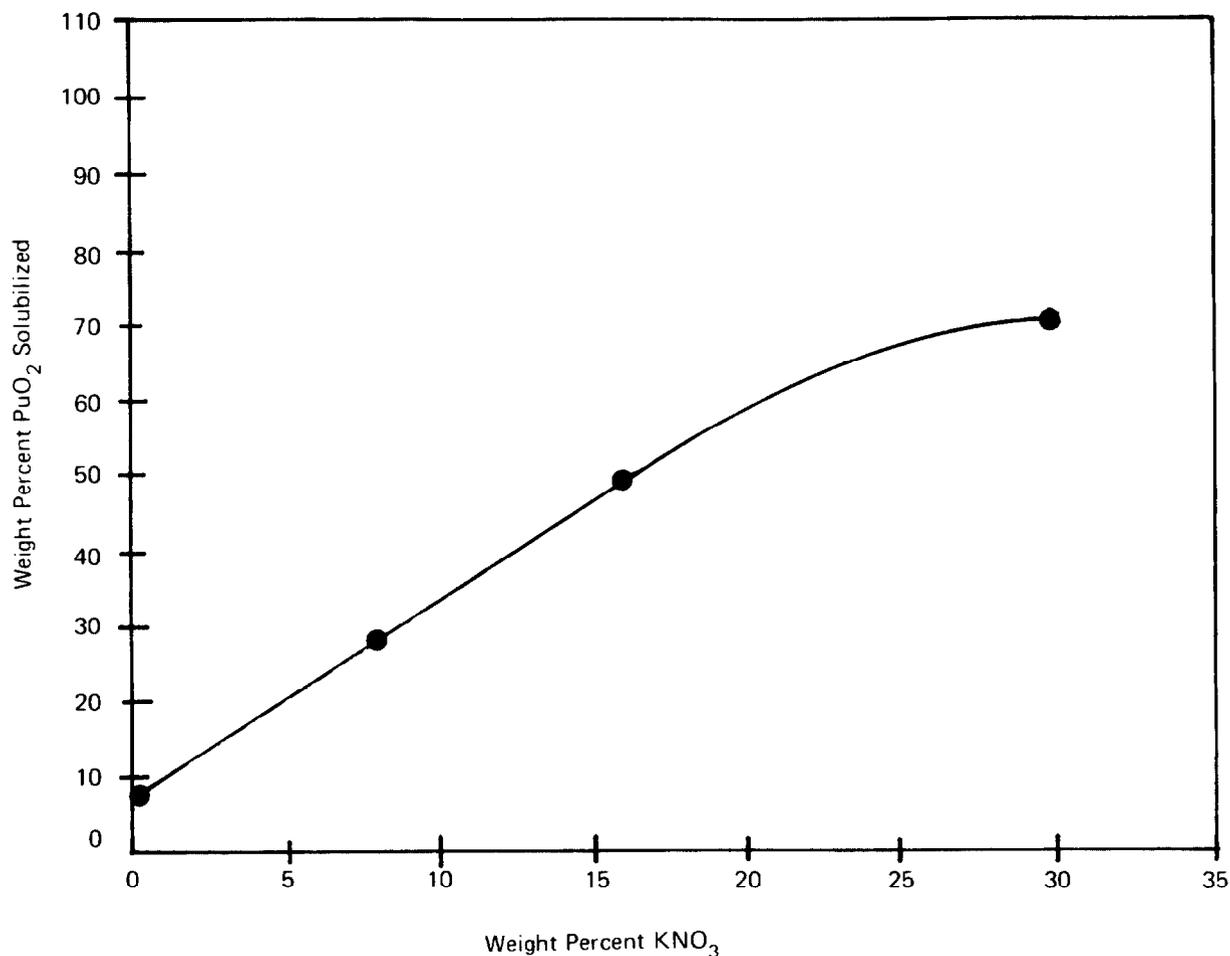


Figure 10 - Effect of salt composition on percent PuO₂ solubilized.

12N HNO₃-0.05N HF-0.01N H₂SO₄. It shows that the dissolution reaction is rapid, with approximately 90% of the PuO₂ dissolved in 4 hr.

Larger samples (40 g) of actual HEPA filter were treated with 12N HNO₃-0.05N HF-0.01N H₂SO₄. These experiments were conducted similarly to those previously described (using 3-g samples) except that different ratios of acid volume (liters) to plutonium-238 weight (grams) were used. Also the filter media were processed through several successive, identical stages in an attempt to obtain the greatest overall decontamination factor (see Table VI and Figure 12). As can be seen in Figure 6, a value of 0.67 for the ratio of acid volume:PuO₂ weight yields a 99.35% dissolution in five stages. On the other hand, a volume:weight ratio of 2.35 produces a 99.53% dissolution in three stages. Thus the decontamination factor is 15⁴ for the lower volume:weight ratio and 21³ for the higher ratio. These final dissolution percentages and decontamination factors were determined by calorimetry and gamma counting of the final filter media residues. All other dissolution percentages and decontamination factors were determined by alpha counting of solution samples.

The fact that a higher volume of acid:weight of PuO₂ ratio produces a higher dissolution is explained as follows: The HF in the

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Table V - Dissolution of Small Samples of Actual HEPA Filters

Acid Test Number	Reagent Composition	Time Heated (hr)	PuO ₂ Dissolved (wt %)	Acid/Pu-238 Ratio (liters/grams)
3265-3A	4N HNO ₃ -0.1M CAN	3-1/2	103.2	1.84
3265-3B	4N HNO ₃ -0.1M CAN	5	103.3	1.84
3265-3C	4N HNO ₃ -0.1M CAN	11-3/4	81.9	1.84
3265-3D	4N HNO ₃ -0.1M CAN	14-3/4	81.5	1.84
3265-4A	12N HNO ₃ -0.05N HF-0.01N H ₂ SO ₄	3-1/2	96.9	1.83
3265-4B	12N HNO ₃ -0.05N HF-0.01N H ₂ SO ₄	5	89.1	1.83
3265-4C	12N HNO ₃ -0.05N HF-0.01N H ₂ SO ₄	12-1/2	93.9	1.83

*Abbreviation for ceric ammonium nitrate - (NH₄)₂ Ce(NO₃)₆.

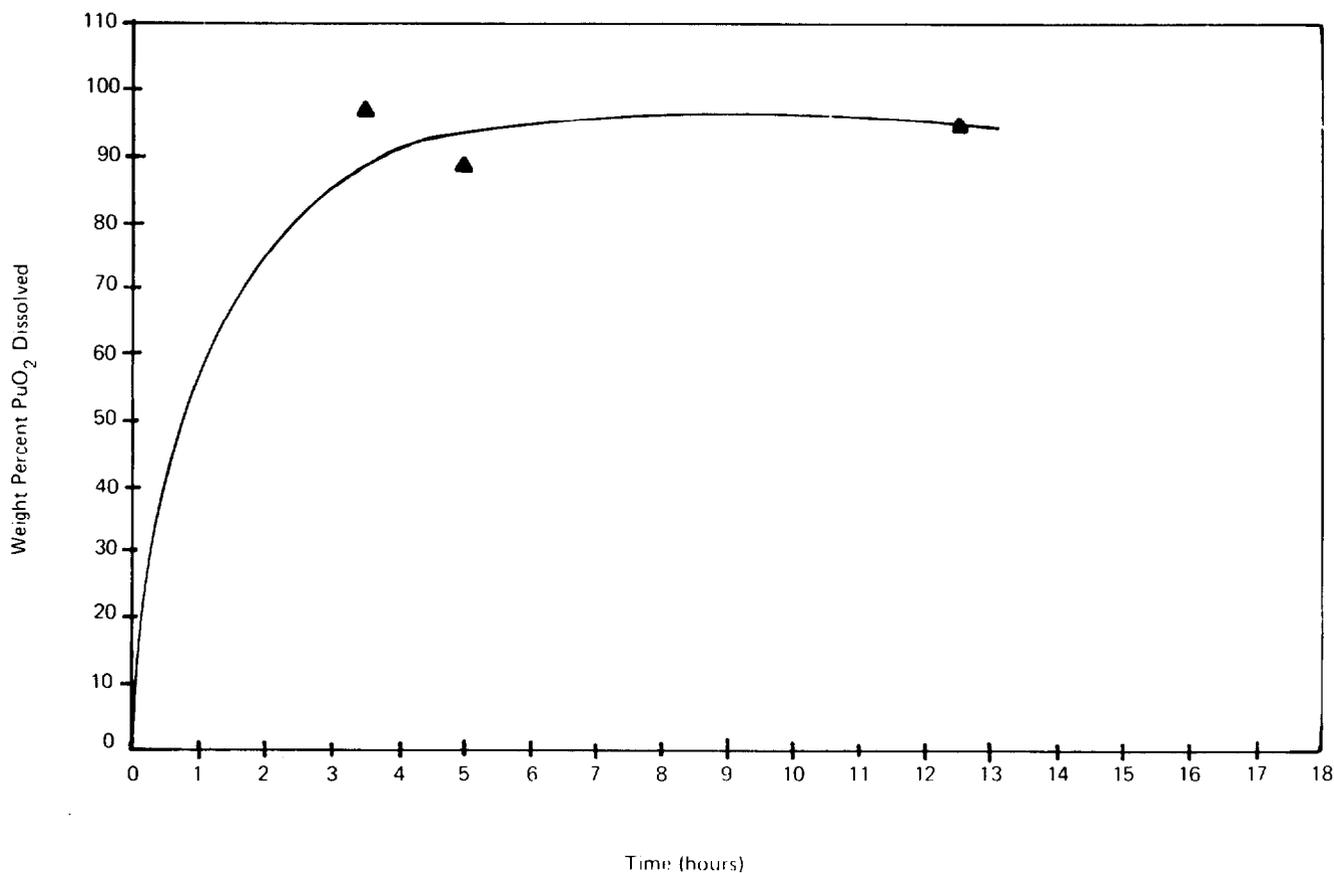


Figure 11 - Dissolution rate of PuO₂ in 12N HNO₃-0.05N HF-0.01N H₂SO₄.

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Table VI - 12N HNO₃-0.05N HF-0.01N H₂SO₄
Dissolution of Large Samples of Actual HEPA Filters

Acid Test Number	Stage	Cumulative PuO ₂ Dissolved (wt %)	Acid/Pu-238 Ratio (liters/g)	Time Heated (hr)
3265-7-1	1	84.9	0.67	3-1/2
3265-7-2	2	88.9	0.67	5-1/4
3265-7-3	3	93.3	0.67	5-3/4
3265-7-4	4	95.8	0.67	7-1/4
3265-7-5	5	99.35	0.67	6-1/2
3265-9-1	1	94.2	2.35	10-1/2
3265-9-2	2	97.6	2.35	10-3/4
3265-9-3	3	99.53	2.35	7

leaching solution is reacting with both PuO₂ particles and glass present in the filter media. When a small amount of leachant is added to a relatively large amount of contaminated filter media, the fluoride ion concentration of the solution is quickly depleted as SiF₄ gas is produced. The remaining HF is not sufficient to dissolve 90% of the PuO₂ present, as desired. Conversely, if a large amount of leachant is added to the same amount of contaminated filter media, there is sufficient HF present to react with the glass and dissolve 90% (or greater) of the PuO₂. What is needed, therefore, is a larger amount of HF in the first stage of the process. This could be accomplished using a larger volume of 12N HNO₃-0.05N HF-0.01N H₂SO₄ or a higher concentration of HF in the acid mixture. For the subsequent stages, an HF concentration of 0.05N should be sufficient since most of the glass has reacted.

Dissolution of PuO₂* in 4N HNO₃-0.15M (NH₄)₂Ce(NO₃)₆

A contaminated filter media sample was prepared by mixing 0.16 g of plutonium dioxide with 3 g of shredded filter media. This sample was placed into a glass beaker containing 250 ml of 4N HNO₃-0.15M ceric ammonium nitrate. The beaker and contents were heated to boiling. Samples were withdrawn periodically and analyzed for plutonium concentration. The solution volume and concentration were kept constant by adding acid of proper concentration in order to replace evaporated acid. Figure 13 shows the effect of Ce⁺⁴ concentration on plutonium dioxide dissolution rate. The 4N HNO₃-0.10M (NH₄)₂Ce(NO₃)₆

*The PuO₂ was a fine powder, having a composition of 80 wt% plutonium-238, 16% plutonium-239, 2.5% plutonium-240, 0.8% plutonium-241, 0.2% plutonium-242, plus small amounts of other actinides, calcined at 950°C.

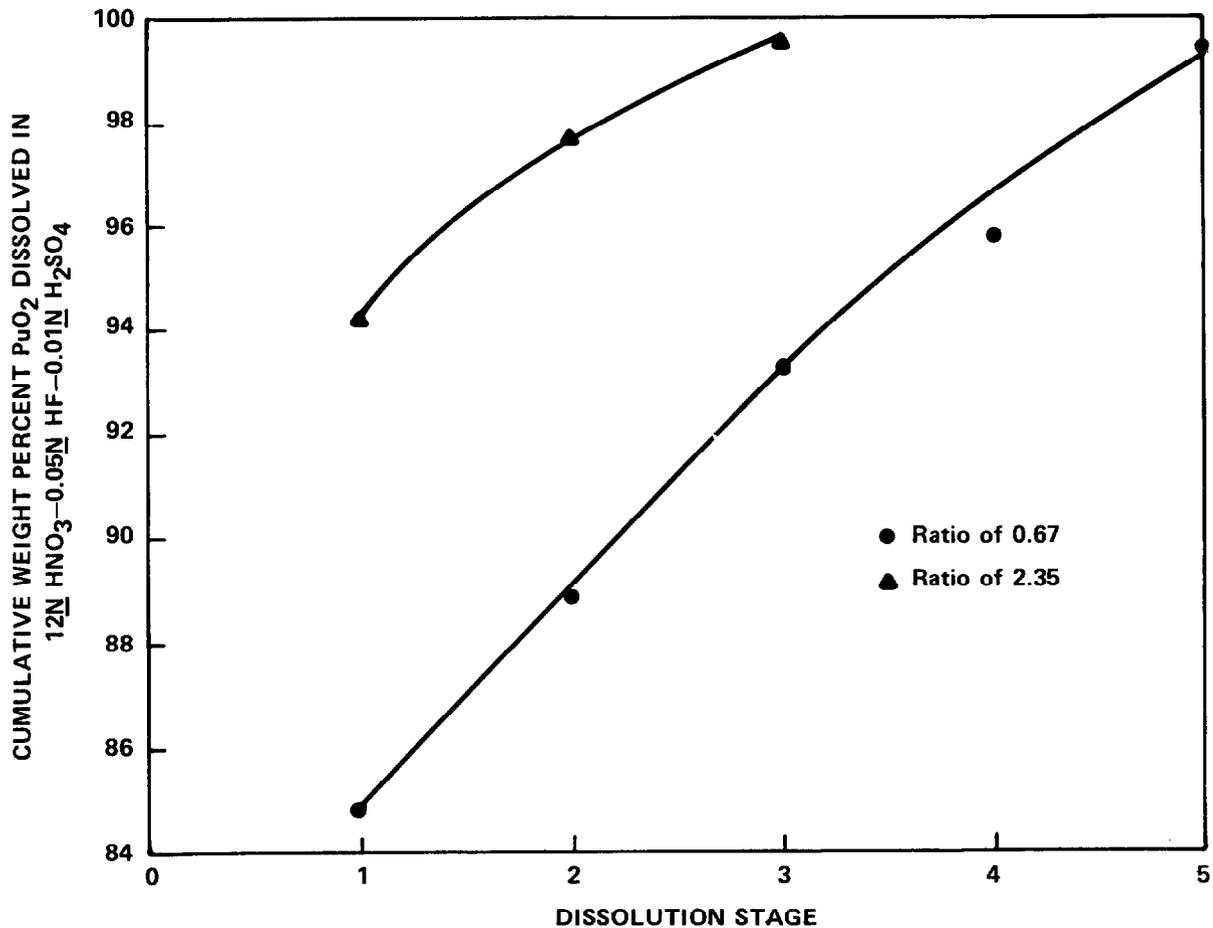


Figure 12 - Effect of acid volume:PuO₂ weight ratio on dissolution percentage.

curve is drawn from data contained in Figure 4. It should be noted that a 50% increase in the Ce⁴⁺ concentration results in a 10% increase in weight percent plutonium dissolved.

Americium-241 and Plutonium-239* Mixed Oxide Dissolution Studies

Contaminated filter media was prepared by mixing AmO₂-PuO₂ powder with shredded filter media. Small samples of this mixture were then treated with various leaching agents, including 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆, 12N HNO₃-0.05N HF-0.01N H₂SO₄, and 8N HNO₃. A sample of filter media was placed in a beaker, 250 ml of the leaching agent was added, and the temperature was increased to the boiling point of the solution. Samples were withdrawn periodically and filtered through a 4-5 μm glass filter. The americium-241 and plutonium-239 concentrations were determined by alpha spectrometry. The solution volume and concentration were kept constant by adding acid of the proper concentration to replace evaporated acid. As can be seen in Table VII, the 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆ was an effective leaching agent, dissolving

* This material was approximately three parts plutonium-239 oxide mixed with one part americium-241 oxide by weight. It had been fired for 2 hr at 1000°C.

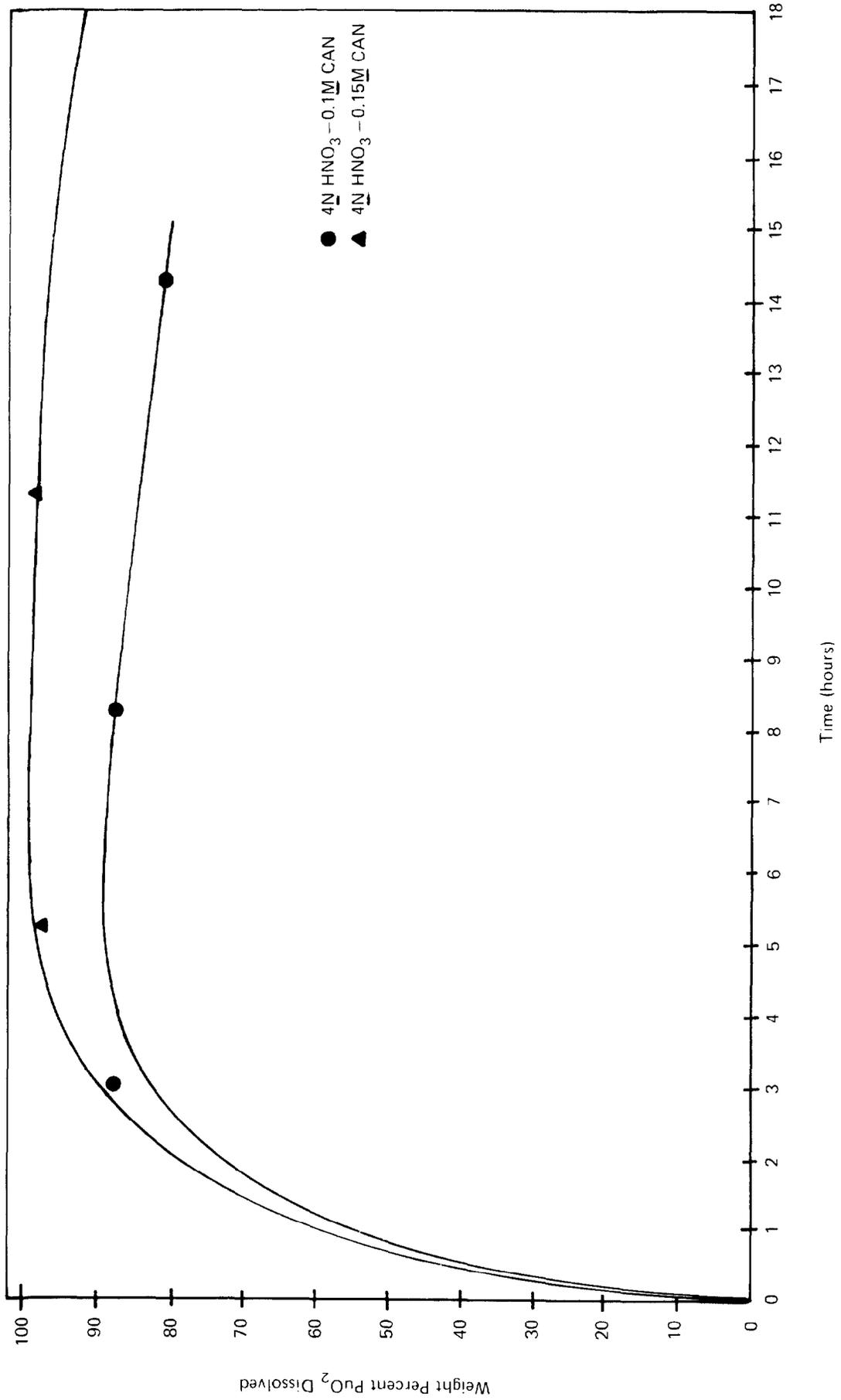


Figure 13 - Effect of ceric ammonium nitrate concentration on PuO₂ dissolution rate.

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Table VII - AmO₂-PuO₂ DISSOLUTION TESTS

Acid Test Number	Reagent Composition	Time Heated (hr)	PuO ₂ Dissolved (wt%)	AmO ₂ Dissolved (wt%)
Am-Pu-1A	4N HNO ₃ -0.1M CAN ^a	5 3/4	72.9	63.2
Am-Pu-1B	4N HNO ₃ -0.1M CAN ^a	11 3/4	93.7	111.8
Am-Pu-2A	12N HNO ₃ -0.05N HF- 0.01N H ₂ SO ₄	5 3/4	68.9	72.4
Am-Pu-2B	"	11 1/4	80.2	90.4
Am-Pu-4A	8N HNO ₃	7 1/4	37.2	88.9
Am-Pu-4B	"	13 1/2	36.3	96.7

^aCAN is an acronym for ceric ammonium nitrate.

greater than 90% of both AmO₂ and PuO₂ in 11-3/4 hr. On the other hand, the 8N HNO₃ was not a satisfactory leaching agent since only 36% of the PuO₂ dissolved in 13 1/2 hr, although 97% of the AmO₂ dissolved during this time (see Figure 14). The mixture of HNO₃-HF-H₂SO₄ was effective in dissolving the AmO₂ (90% in 11-1/4 hr) but not as effective in dissolving the PuO₂ (80% in 11-1/4 hr). Of the three reagents tested, the 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆ is the recommended leaching agent for mixtures of AmO₂-PuO₂.

Uranium-233 Oxide Dissolution Studies

These studies were conducted using U₃O₈ mixed with filter media. The oxide had been fired at 950°C for 2 hr. A small sample of the prepared filter media (approximately 3.2 g) was placed into a beaker containing 250 ml of boiling reagent. The reagents used were 8N HNO₃, 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆, and 12N HNO₃-0.05N HF-0.01N H₂SO₄. Samples were withdrawn periodically for alpha analysis. All samples were filtered through 4-5 μm glass filters. The solution volume and concentration were kept constant by adding acid of the proper concentration to replace evaporated acid. As can be seen from Table VIII, in all three cases the dissolution was very rapid and exceeded 90%. For example, the dissolution rate in 8N HNO₃ is depicted in Figure 16. All three reagents would be acceptable dissolution reagents for uranium-233 oxide.

Sodium Carbonate Fusion of PuO₂*

These studies were conducted in order to determine the effect of temperature on fusion efficiency. Small samples were prepared by mixing 64 mg of PuO₂ with 2.25 g of Na₂CO₃. The PuO₂-Na₂CO₃ mixture was then placed into a platinum crucible and contents were then heated to the desired operating temperature (1200°C and 1300°C) and maintained

*The PuO₂ was a fine powder having a composition of 80 wt% plutonium-238, 16 wt% plutonium-239, 2.5 wt% plutonium-240, 0.8 wt% plutonium-241, 0.2 wt% plutonium-242, plus small amounts of other actinides. It was calcined for 2 hr at 950°C.

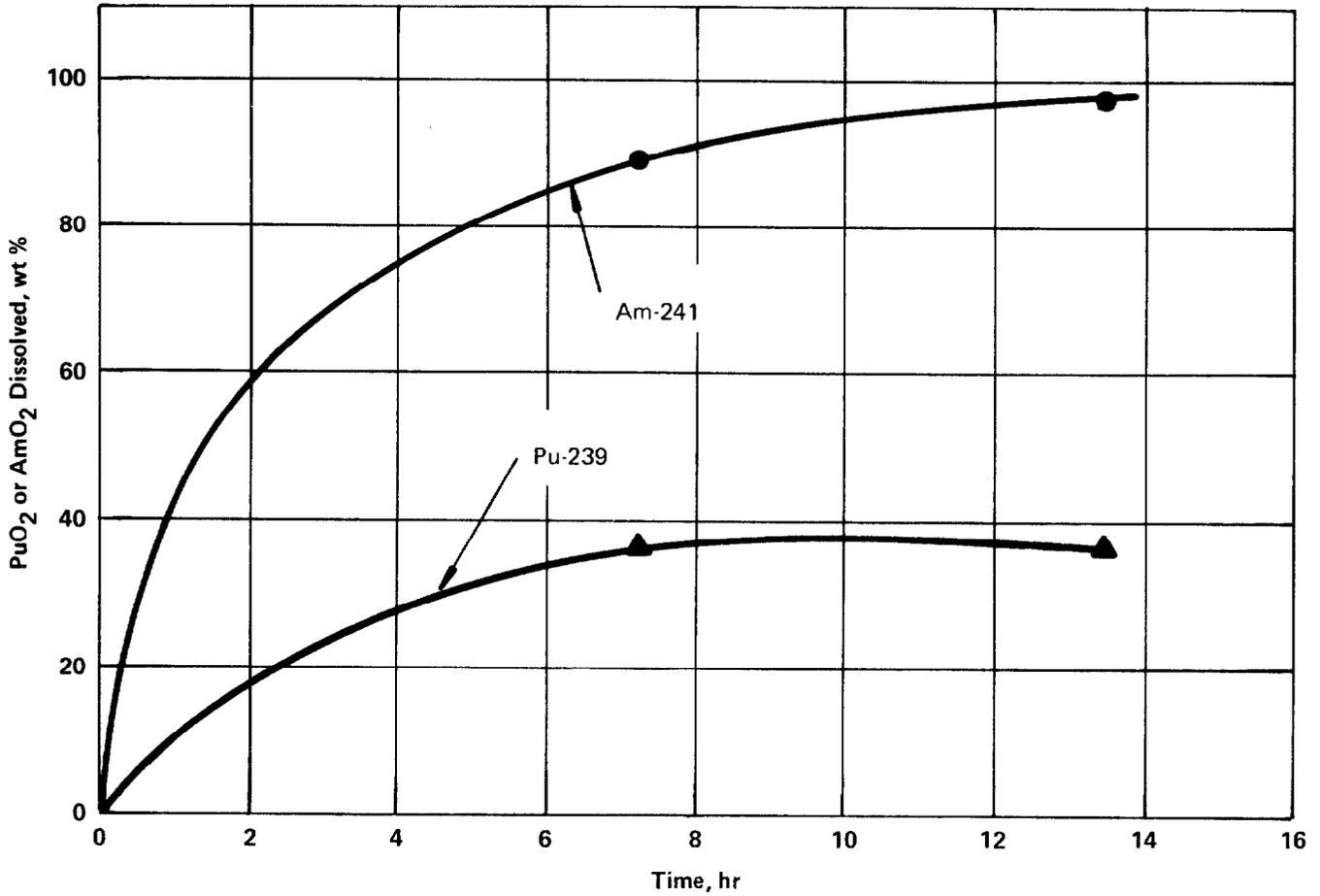


Figure 14 - Dissolution rate of Am²⁴¹-Pu²³⁹ oxides in 8N HNO₃.

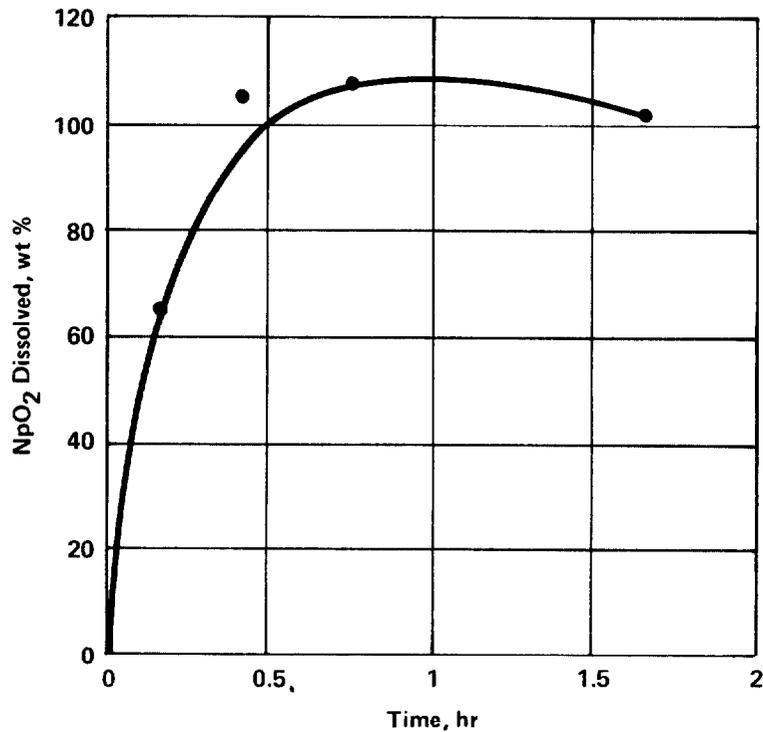


Figure 15 - Dissolution rate of U₃O₈ in 8N HNO₃-0.1N HF

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Table VIII- U-233 OXIDE DISSOLUTION TESTS

Acid Test Number	Reagent Composition	Time Heated (min)	U-233 Oxide Dissolved (wt%)
U-3-1	8N HNO ₃	5	97.4
U-3-2	"	20	106.1
U-3-3	"	65	93.8
U-3-4	"	255	95.9
U-4-1	4N HNO ₃ -0.1M CAN ^a	5	89.7
U-4-2	"	15	104.8
U-4-3	"	60	95.2
U-4-4	"	360	95.7
U-5-1	12N HNO ₃ -0.05N HF- 0.01N H ₂ SO ₄	5	100.9
U-5-2	"	15	106.2
U-5-3	"	50	100.1
U-5-4	"	240	102.5

^aCAN is an acronym for ceric ammonium nitrate.

there for 1 hr. The crucible and melt were cooled to room temperature, and the melt was removed from the crucible and dissolved in 4N HNO₃. As can be seen in Table IX, there is not a significant difference (4.6%) in the two dissolution percentages and it is probable that the difference is due to experimental error. Therefore, it can be concluded that within the temperature range under investigation, the fusion efficiency is approximately 58% (average value).

Neptunium-237 Oxide Dissolution Studies

The NpO₂ was fired for 2 hr at 950°C. Samples of contaminated filter media were prepared by mixing 0.2 g of NpO₂ with 3 g of filter media. The procedure was identical to that used in the uranium-233 oxide dissolution studies. The following three leaching reagents were tested: 8N HNO₃, 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆, and 12N HNO₃-0.05N HF-0.01N H₂SO₄. Table X lists the results from these experiments. As can be seen, the 8N HNO₃ was unsuccessful in dissolving the NpO₂ rapidly with only 32% solubilized in 8 hr. On the other hand, each of the other two reagents dissolved the NpO₂ rapidly with greater than 90% dissolved after 1/2 hr of boiling (see Figure 15 for dissolution percentage as a function of time). Therefore, both the 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆ and the 12N HNO₃-0.05N HF-0.01N H₂SO₄ acid mixtures would be recommended for dissolution of NpO₂ from filter media.

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Table IX - Na₂CO₃ FUSIONS OF PuO₂

<u>Fusion Test Number</u>	<u>Time Heated (hr)</u>	<u>Operating Temperature (°C)</u>	<u>PuO₂ Solubilized (wt%)</u>	<u>Dissolution Reagent</u>
Pu-44	1	1300°C	56.4	4N HNO ₃
Pu-45	1	1200°C	61.0	4N HNO ₃

Table X - Np-237 OXIDE DISSOLUTION TESTS

<u>Acid Test Number</u>	<u>Reagent Composition</u>	<u>Time Heated (min)</u>	<u>NpO₂ Dissolved (wt%)</u>
Np-3-1	8N HNO ₃	10	1.3
Np-3-2	"	25	1.9
Np-3-3	"	45	4.1
Np-3-4	"	75	6.1
Np-3-5	"	480	32.3
Np-4-1	4N HNO ₃ -0.1M CAN ^a	10	93.7
Np-4-2	"	25	97.2
Np-4-3	"	40	88.9
Np-4-4	"	60	89.1
Np-5-1	12N HNO ₃ -0.05N HF-0.01N H ₂ SO ₄	10	64.7
Np-5-2	"	25	105.2
Np-5-3	"	45	107.4
Np-5-4	"	100	102.3

^aCAN is an acronym for ceric ammonium nitrate.

Curium-244* Oxide Dissolution Studies

Contaminated filter media were prepared by mixing 13 mg of CmO₂ with 3 g of filter media. The experimental procedures were identical to those used for the uranium-233 dissolution studies. The two leaching reagents tested were 12N HNO₃-0.05N HF and 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆. The results of these tests, listed in Table XI, show that both reagents dissolved the CmO₂ very rapidly (essentially 100% dissolution in 10 min). Figure 17 shows the dissolution rate for 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆. Both reagents are recommended for dissolution of CmO₂ from contaminated filter media.

*This was a powder which was approximately 94% curium-244 and 4% curium-246, the remainder being curium-245, -247, and -248.

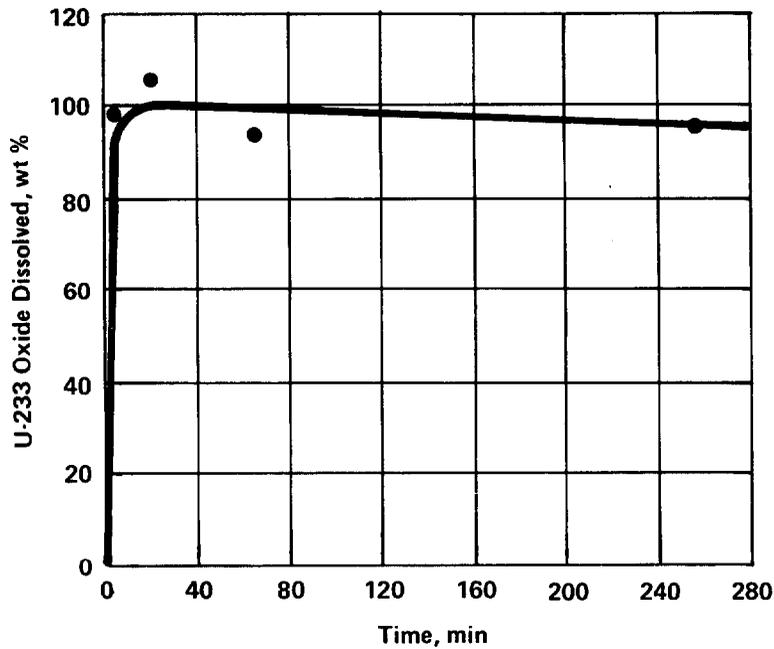


Figure 16- Dissolution rate of U-233 oxide in 8N HNO₃.

Table XI - Cm-244 OXIDE DISSOLUTION TESTS

Acid Test Number	Reagent Composition	Time Heated (min)	CmO ₂ Dissolved (wt%)
Cm-2A	12N HNO ₃ -0.05N HF	10	103.3
Cm-2B	"	25	100.8
Cm-2C	"	60	105.0
Cm-2D	"	120	99.1
Cm-2E	"	180	104.5
Cm-2F	"	540	93.3
Cm-2G	"	540	99.0
Cm-3A	4N HNO ₃ -0.1M CAN ^a	10	112.1
Cm-3B	"	25	110.0
Cm-3C	"	60	109.0
Cm-3D	"	120	107.6
Cm-3E	"	330	110.5

^aCAN is an acronym for ceric ammonium nitrate.

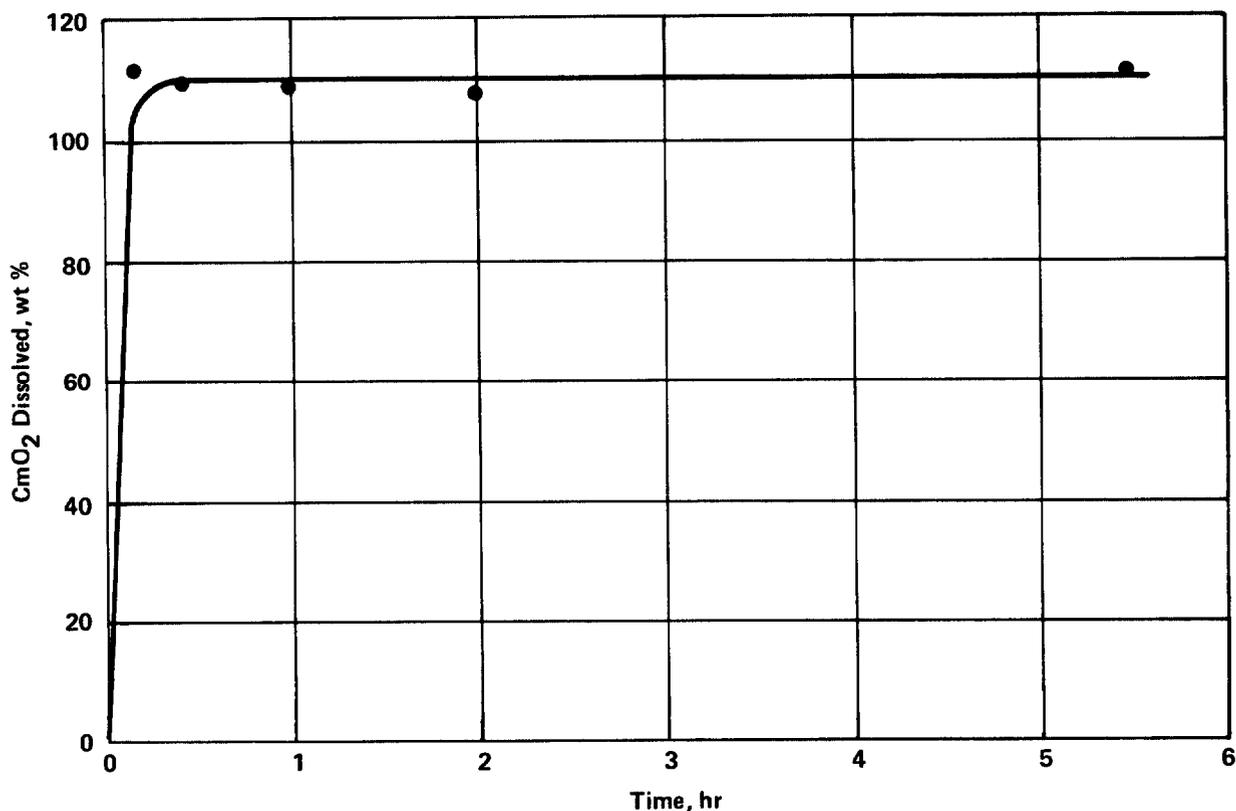


Figure 17 - Dissolution rate of CmO_2 in $4\text{N HNO}_3-0.1\text{M } (\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$.

Thorium-232 Oxide Dissolution Tests

Contaminated filter media were prepared by mixing 0.24 g of ThO_2 with 3 g of filter media. The experimental procedure was identical to that used in the uranium-233 oxide studies. The ThO_2 was a fine powder which had been fired at 600°C . The purpose of the experiment was to determine whether ThO_2 would dissolve in $4\text{N HNO}_3-0.1\text{M } (\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$ similarly to other actinide oxides previously tested (PuO_2 , CmO_2 , AmO_2 , U_3O_8 , and NpO_2). As shown by the data in Table XII and Figure 18, the dissolution rate was slow at first (0-7 hr), rapid from 7 to 9 hr, and slow again from 9 to 20 hr. The reason for this is not known at this time, but it is thought that it took 7 hr to attack the surface of the ThO_2 and open the pores so that rapid dissolution could begin. It therefore appears that $4\text{N HNO}_3-0.1\text{M } (\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$ would be an acceptable leaching agent for ThO_2 from filter media, although dissolution time is longer than for other actinides.

A point of interest is that in a similar experiment using just ThO_2 (no filter media), the dissolution percentage obtained was only 25% after 20 hr of boiling. The reason for the difference in dissolution percentages (25% versus 100%) is that the filter media contains 0.14 wt% F^{-1} . This small amount of fluoride ion has been found to be sufficient to catalyze the dissolution reaction.

Dissolution of PuO_2 from Actual Glovebox Filters

These tests were conducted using samples of filter media removed from actual spent glovebox filters. The PuO_2 on the filters was the same type as that used in the fusion experiments.

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Table XII - ThO₂ DISSOLUTION DATA

Acid Test Number	Reagent Composition	Time Heated (min)	ThO ₂ Dissolved (wt%)
Th-1	4N HNO ₃ -0.1M CAN ^a	5	0.6
Th-2	"	10	0.4
Th-3	"	20	1.4
Th-4	"	35	2.4
Th-5	"	60	5.1
Th-6	"	180	4.7
Th-7	"	300	12.5
Th-8	"	440	31.7
Th-9	"	440	29.3
Th-10	"	470	42.1
Th-11	"	545	82.3
Th-12	"	905	100.5
Th-13	"	1205	104.3

^aCAN is an acronym for ceric ammonium nitrate.

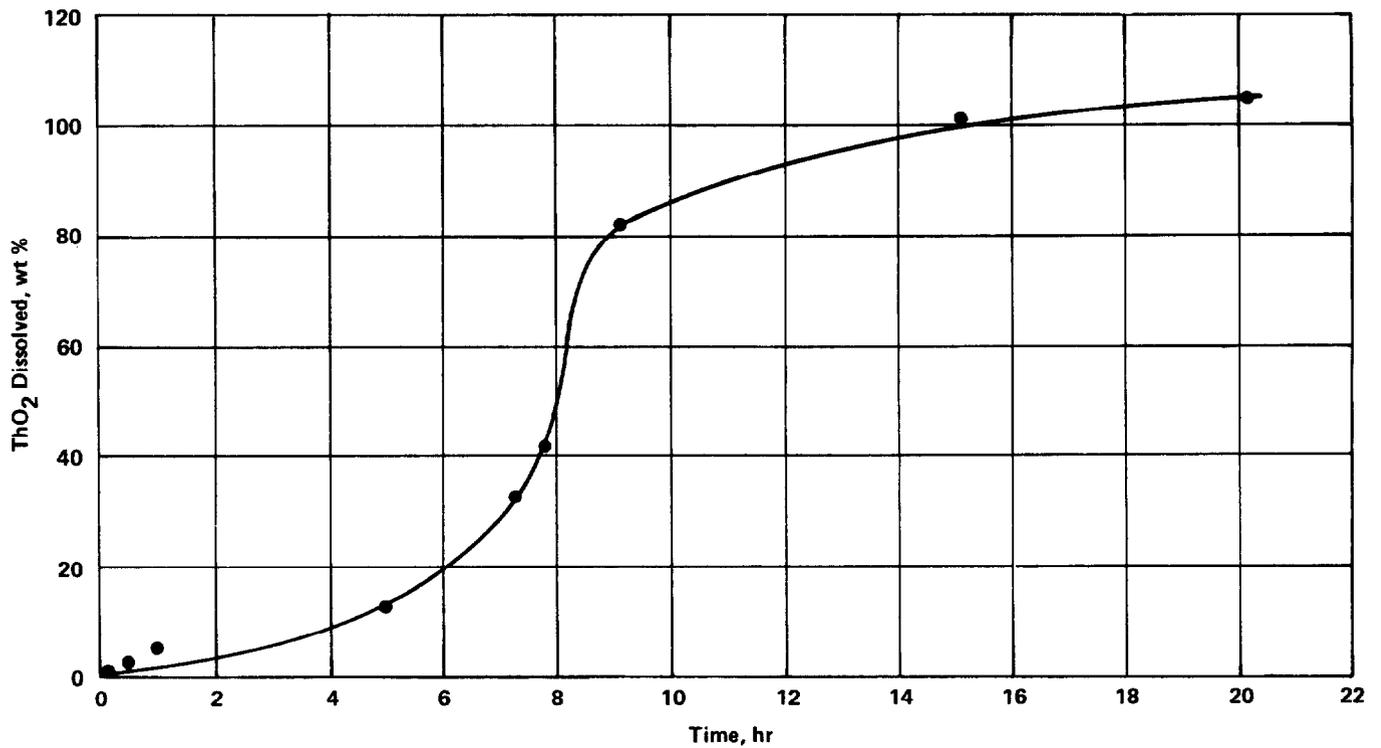


Figure 18 - Wt % ThO₂ dissolved in 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆.

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Experiment A

A 9.5 g sample of filter media containing 0.18 g of PuO_2 was dissolved in a six-stage dissolution procedure. The leaching reagent in the first five stages was 170 ml of 4N HNO_3 -0.23M $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$, and the sixth stage contained 12N HNO_3 -0.1N HF. The 12N HNO_3 -0.1N HF was used because the 4N HNO_3 -0.23M $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$ had proved ineffective in the fifth stage of dissolution. The volume of 12N HNO_3 -0.1N HF used was 250 ml and all studies were done at boiling temperature. The solution was sampled periodically for plutonium concentration, and all samples were filtered through 4-5 μm glass filters. When the dissolution rate for a particular stage had become very slow (as indicated by α analysis of solution), the filter media were separated from the leaching solution by filtration and placed in the next stage of dissolution.

Results for this experiment are given in Table XIII. Figures 19 through 21 show the dissolution rate for each stage, and Figure 22 shows the cumulative PuO_2 dissolution percentage for the six-stage process. It should be noted that final dissolution percentage was 99.88% (a contamination factor of 833). This was determined by gamma ray spectrometry of the residual filter media and agrees well with the final value as determined by alpha counting of the solution samples (99.80%). It should also be noted that the 12N HNO_3 -0.1N HF solution used in the sixth stage was more than twice as effective as the 4N HNO_3 -0.23 M $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$ used in the fifth stage (0.079% dissolved versus 0.031%). The reason for this is not understood at this time. More studies will be done to investigate this phenomenon.

Experiment B

A 25.6 g sample of filter media containing 0.31 g of PuO_2 was dissolved in a four-stage dissolution process. The leaching reagent used in the test was 460 ml of 4N HNO_3 -0.3M $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$. The experimental procedure was identical to that used in Experiment A of this section. The results for this experiment are given in Table XIV; Figure 23 shows the cumulative PuO_2 dissolution percentage for the four stage process. The dissolution percentages in Table XIV were determined by alpha counting, but the cumulative percentages used in Figure 23 were based on a final gamma spectrometry value of 98.24%. This latter value compares well with the value of 97.62% obtained by alpha counting. The gamma value is, however, considered more accurate than the total obtained by simple addition of the individual alpha count values. The decontamination factor obtained using this gamma value is 88.5. The decontamination factor obtained after four stages in Experiment A of this section was 417.

A logical question would be: Why was the decontamination factor less when more ceric ammonium nitrate per mole of PuO_2 was used? It is known that the 25.6 g sample filter media contained a greater percentage of pack-to-frame sealant* than the 9.5-g sample. It is also

*This is the material that is used to seal the asbestos-glass filter pack to the wooden frame. It is a fire retardant polyurethane foam and a rubber base adhesive.

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Table XIII- PuO₂ DISSOLUTION DATA (9.5-g sample)

Acid Test Number	Reagent Composition	Time Heated (min)	PuO ₂ Dissolved (wt%)	Process Stage
256-1 A	4N HNO ₃ -0.23M CAN ^a	10	82.0	1
256-2 A	"	20	79.5	1
256-3 A	"	40	87.3	1
256-4 A	"	60	80.9	1
256-5 A	"	120	85.8	1
256-6 A	"	210	90.6	1
256-1 B	"	10	0.41	2
256-2 B	"	20	1.17	2
256-3 B	"	45	2.50	2
256-4 B	"	65	2.97	2
256-5 B	"	120	4.75	2
256-6 B	"	360	7.30	2
256-7 B	"	510	7.47	2
256-1 C	"	30	0.29	3
256-2 C	"	75	0.55	3
256-3 C	"	135	0.87	3
256-4 C	"	315	1.06	3
256-5 C	"	435	1.09	3
256-6 C	"	675	1.31	3
256-7 C	"	675	1.39	3
256-1 D	"	20	0.029	4
256-2 D	"	45	0.063	4
256-3 D	"	60	0.079	4
256-4 D	"	135	0.139	4
256-5 D	"	330	0.213	4
256-6 D	"	645	0.222	4
256-7 D	"	645	0.228	4
256-1 E	"	15	0.011	5
256-2 E	"	30	0.012	5
256-3 E	"	60	0.015	5
256-4 E	"	135	0.023	5
256-5 E	"	195	0.025	5
256-6 E	"	465	0.029	5
256-7 E	"	600	0.031	5
256-8 E	"	780	0.032	5
256-9 E	"	780	0.025	5
256-1 F	12N HNO ₃ -0.1N HF	30	0.016	6
256-2 F	"	60	0.030	6
256-3 F	"	240	0.047	6
256-4 F	"	585	0.079	6
256-5 F	"	945	0.075	6

^aCAN is an acronym for (NH₄)₂Ce(NO₃)₆.

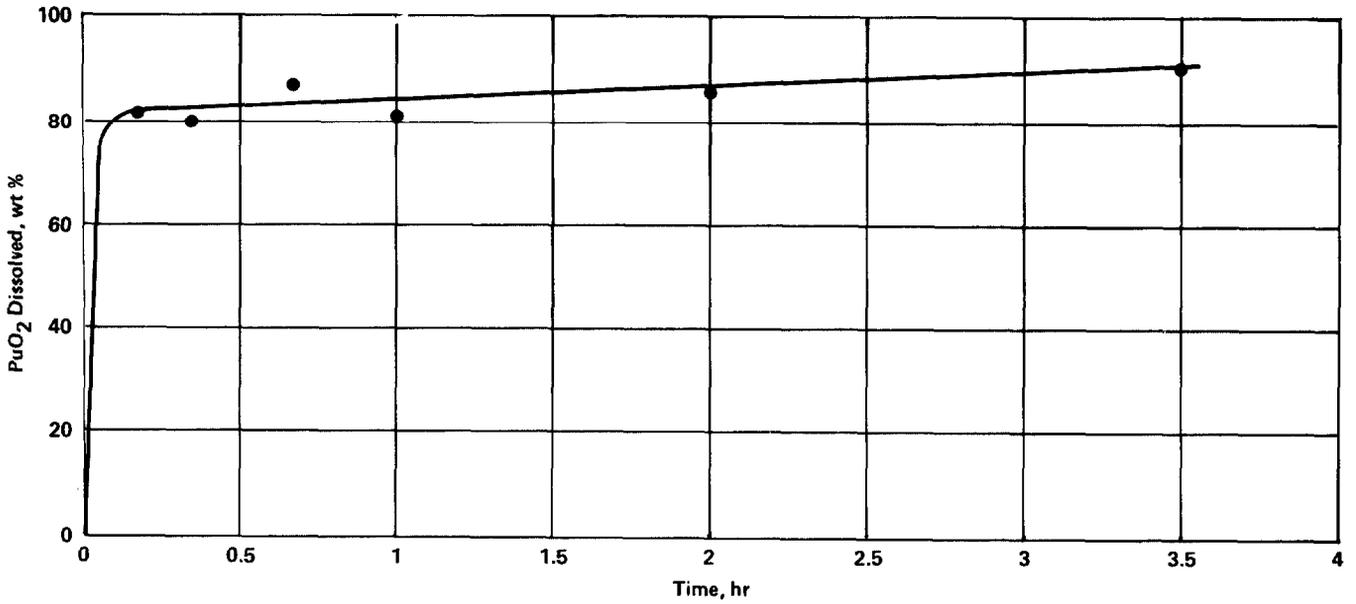


Figure 19 - Dissolution rate of PuO₂ from filter media (1st stage).

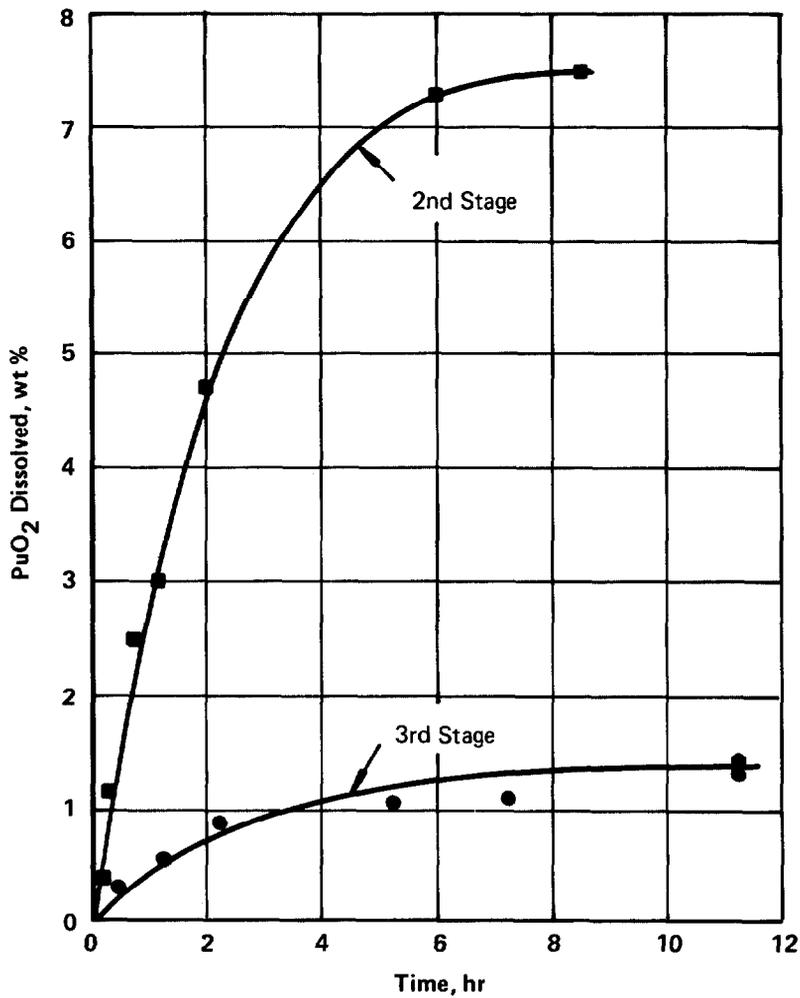


Figure 20 - Dissolution of PuO₂ from filter media (2nd and 3rd stage).

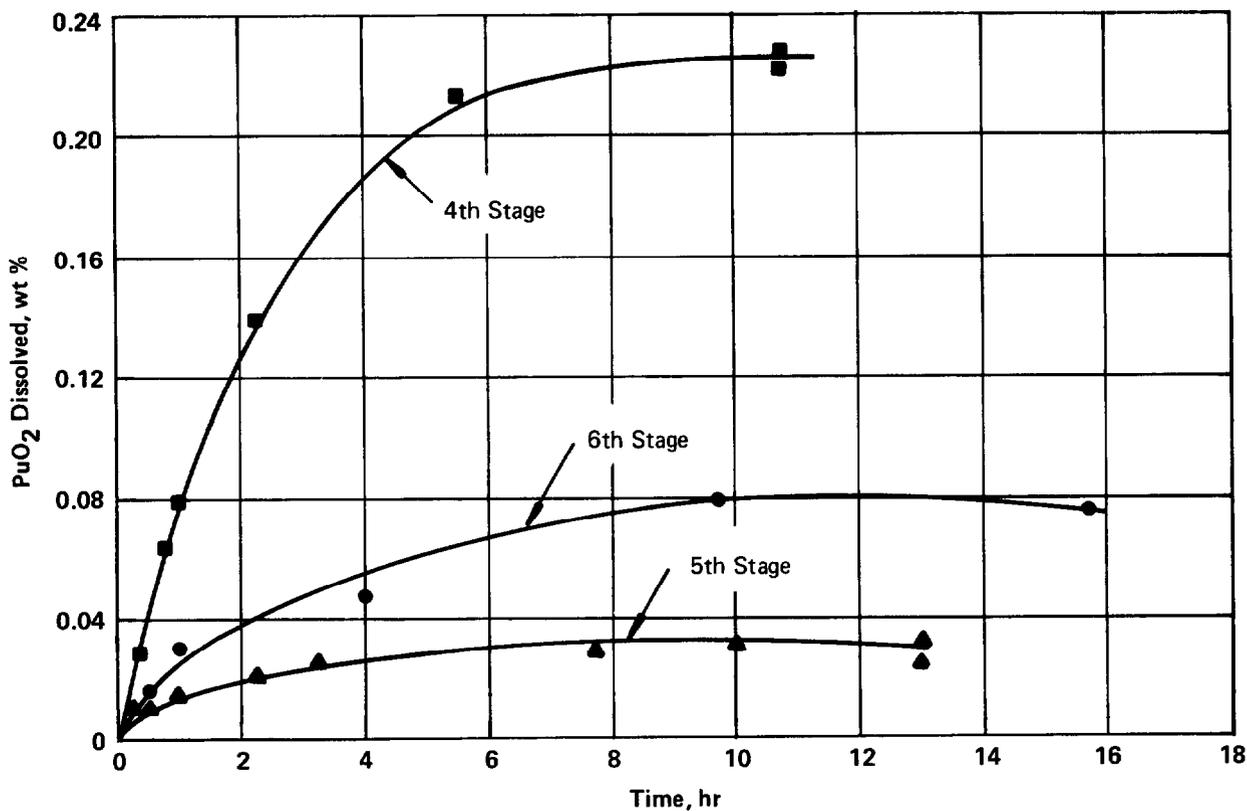


Figure 21 - Dissolution of PuO₂ from filter media (4th, 5th, and 6th stage).

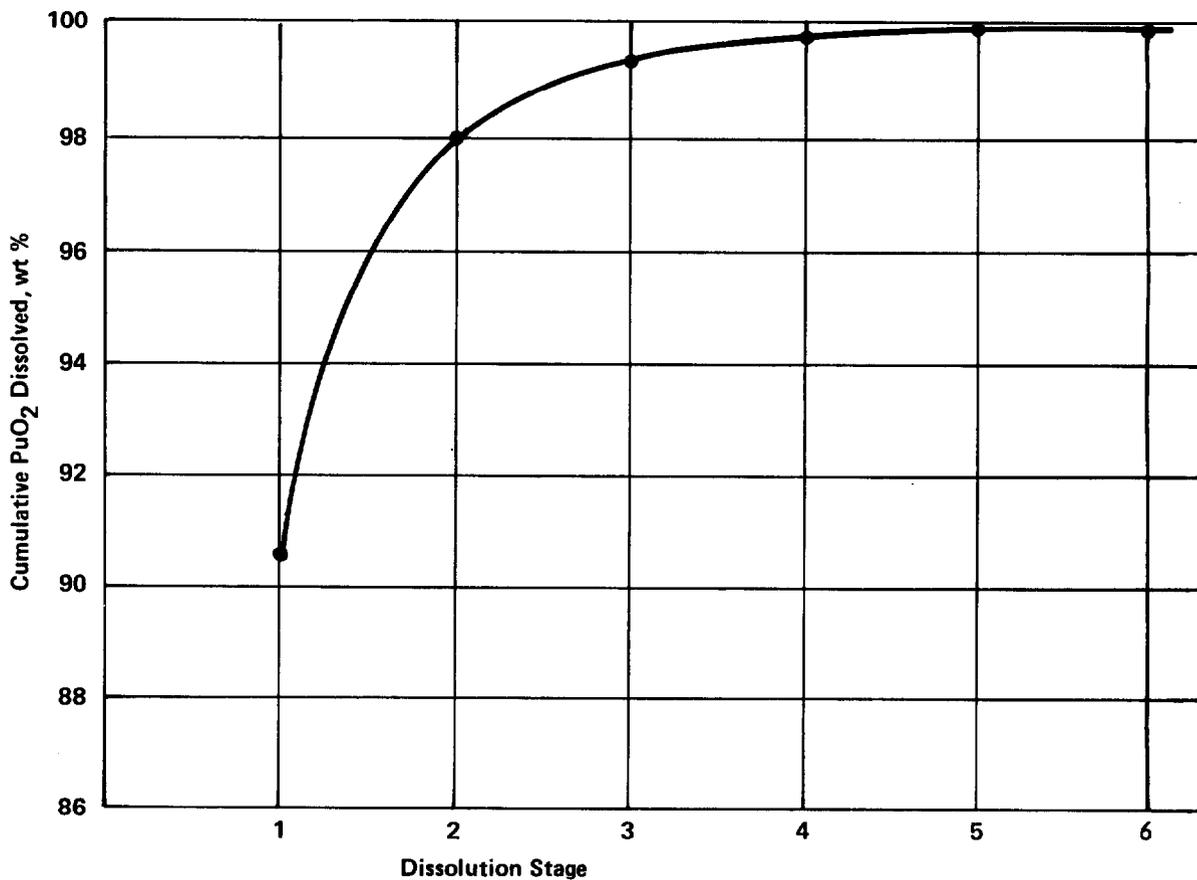


Figure 22 - Cumulative wt % PuO₂ dissolved during six-stage process.

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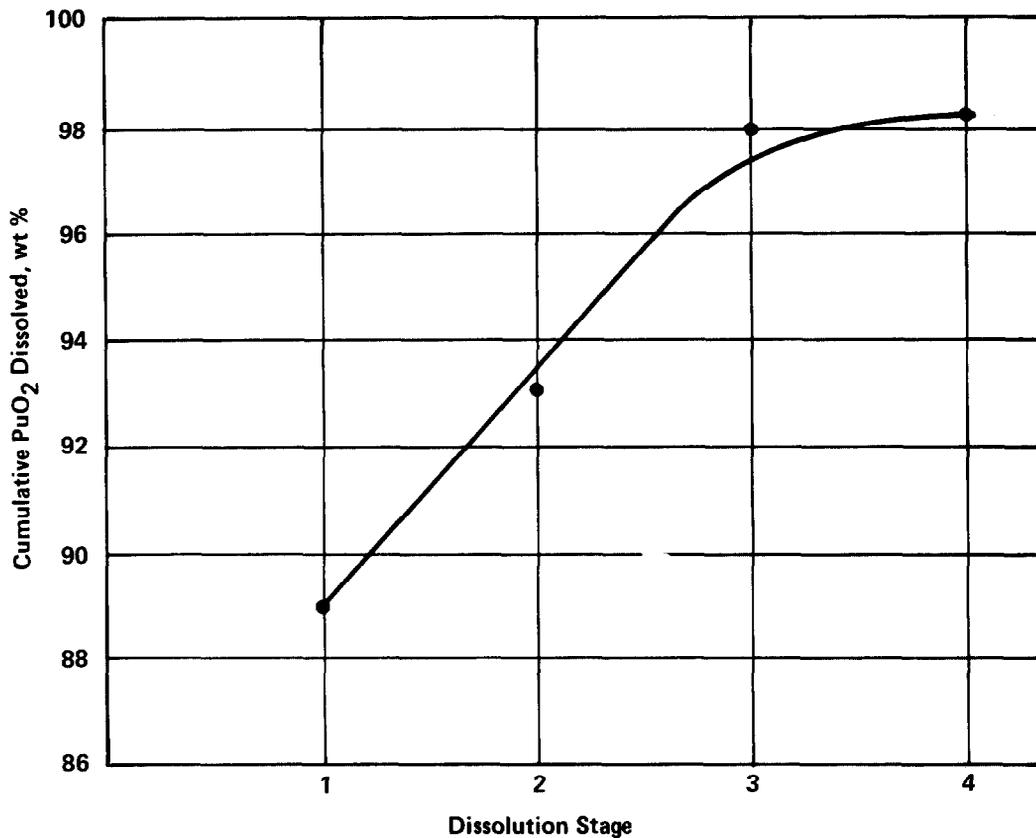


Figure 23 - Cumulative wt % PuO₂ dissolved during four-stage process.

Table XIV- PuO₂ DISSOLUTION DATA (25.6-g sample)

Acid Test Number	Reagent Composition	Stage	Time Heated (hr)	PuO ₂ Dissolved (wt%)	Cumulative PuO ₂ Dissolved (wt%)
12-1	4N HNO ₃ -0.3M CAN ^a	1	6 1/2	88.40	88.40
12-2	"	2	15	4.04	92.44
12-3	"	3	7 1/4	4.91	97.35
12-4	"	4	7	0.27	97.62 ^b

^aCAN is an acronym for ceric ammonium nitrate.

^bGamma ray analysis of remaining filter media indicated that 98.24% of the PuO₂ had been dissolved.

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known (from the color change of the leaching solution) that the Ce^{+4} ions were reduced much more quickly during the dissolution of PuO_2 from the 25.6-g filter media sample. Therefore, it is surmised that the reason for the smaller decontamination factor (smaller percentage dissolved) is that the Ce^{+4} ions were used mainly to oxidize the sealant thus leaving fewer Ce^{+4} ions to oxidize to Pu^{+4} . A simple solution to this problem might be to increase the amount of ceric ammonium nitrate when larger amounts of sealant are present with the filter media.

Summary

Plutonium adsorption isotherms were developed for the $Pu-HNO_3$ and $Pu-HNO_3-HF$ systems which proved that the filter media did indeed adsorb plutonium. The x/m ratio for the $Pu-HNO_3-HF$ system was less than that for the $Pu-HNO_3$ system at any given concentration. Thus, higher decontamination factors can be obtained with HNO_3-HF (assuming each leaching agent is equally effective in the dissolution of plutonium dioxide).

Dissolution tests were conducted on filter media contaminated with PuO_2 . The $HNO_3-H_2SO_4$ mixtures were relatively ineffective as leaching agents (see Figure 3). The best dissolution agents found were mixtures of HNO_3-HF and $HNO_3-HF-H_2SO_4$ in which the HF concentration was 0.05N or greater. These solutions dissolved greater than 90% of the PuO_2 in 15 hr (see Figures 5 and 6). Several tests were performed with $HNO_3-(NH_4)_2Ce(NO_3)_6$ solutions (see Figure 4). It was found that 4N $HNO_3-0.1M (NH_4)_2Ce(NO_3)_6$ was the best of these mixtures with 88% of the PuO_2 dissolved in 3 hr. Addition of $KMnO_4$ to 4N HNO_3 -cerium(III) dissolution mixtures was successful in oxidizing the cerium(III) to cerium(IV) and thus allowing more of the PuO_2 to dissolve.

Several dissolution tests were completed with UO_2 and U_3O_8 contaminated filter media. All leaching agents tested dissolved the uranium oxide rapidly (see Table II). Equally good results were obtained with 8N HNO_3 which dissolved ~100% of the UO_2 or U_3O_8 in 6 hr.

Neither Na_2CO_3 nor $Na_2CO_3-KNO_3$ was satisfactory as a fusion agent for PuO_2 . The greatest percentage of PuO_2 recovered was 71% using a Na_2CO_3-30 wt % KNO_3 salt mixture. Also there are tremendous corrosion problems associated with this process. The platinum crucibles and the furnace used in these experiments were corrosively attacked by the resultant vapors.

The $PuO_2-75\% UO_2$ solid solution dissolved readily in 8N HNO_3 , 12N $HNO_3-0.1N HF$, 12N $HNO_3-0.05N HF-0.01N H_2SO_4$, and 4N $HNO_3-0.1M (NH_4)_2Ce(NO_3)_6$. These would be acceptable leaching agents since greater than 93% of the PuO_2 dissolved in 2 hr at boiling temperatures. Hueda (4) has reported that PuO_2-UO_2 can be dissolved in nitric acid alone (up to 35% PuO_2). Baehr and Dippel (5) reported that $UO_2-15\% PuO_2$ fired at 1600°C will dissolve easily in 14M HNO_3 . These sources substantiate recent findings at Mound Facility.

Small and large samples of actual HEPA glovebox filters (contaminated with PuO_2) were treated with various leaching solutions. For small samples, 12N $HNO_3-0.05N HF-0.01N H_2SO_4$ proved superior to 4N $HNO_3-0.1M (NH_4)_2Ce(NO_3)_6$ with greater than 89% of the PuO_2 dissolved. For larger samples, it was found that an increase in the ratio of acid volume to PuO_2 weight increased the dissolution percentage. Figure 12

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compares a five-stage process (volume/weight ratio of 0.67) with a three-stage process having volume:weight ratio of 2.35. It should be noted that the latter process obtained a higher percentage of dissolution than the former.

Dissolution studies using 4N HNO₃-0.15M ceric ammonium nitrate dissolved greater than 93% of the initial PuO₂. Therefore, 4N HNO₃ in combination with (NH₄)₂Ce(NO₃)₆ is a recommended leaching reagent.

Filter media which were contaminated with americium-241 and plutonium-239 oxides were treated with three different leaching reagents to determine the effectiveness of each reagent in dissolving the actinide oxides. The three reagents used were 8N HNO₃, 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆, and 12N HNO₃-0.05N HF-0.01N H₂SO₄. It was found that the best leaching agent was the 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆ which dissolved greater than 90% of the AmO₂ and the PuO₂.

Another series of tests was conducted to determine the best leaching agent for uranium-233 oxide (U₃O₈). Simulated samples of contaminated filter media were prepared by mixing 0.2 g of U₃O₈ powder with 3 g of shredded filter media. The reagents used were the same three as were used for the AmO₂-PuO₂ studies. It was found that all three reagents were successful in dissolving greater than 90% of the U₃O₈ in a very short time (15 min).

Fusion studies were conducted using Na₂CO₃ and PuO₂. The purpose of these experiments was to determine the effect of temperature on fusion efficiency. It was found that there was not a significant difference (4.6%) in the PuO₂ solubilized within the temperature range under investigation (1200-1300°C).

Experiments were conducted using filter media contaminated with neptunium-237 oxide. The purpose of the tests was to determine which of the three reagents would be successful in dissolving the NpO₂. Results indicate that both the 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆ and the 12N HNO₃-0.05N HF-0.01N H₂SO₄ were successful in dissolving greater than 90% of the NpO₂ in 0.5 hr. The 8N HNO₃ on the other hand solubilized only 32% of the NpO₂ in 8 hr and therefore would not be recommended as a leaching agent for NpO₂.

In other series of experiments, filter media contaminated with curium-244 oxide were dissolved in leaching tests using both 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆ and 12N HNO₃-0.05N HF. Both reagents dissolved essentially 100% of the CmO₂ in approximately 10 min and therefore would be recommended for dissolving CmO₂ from filter media.

Filter media contaminated with thorium-232 oxide were leached with a 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆ solution in an attempt to dissolve the ThO₂. The rate of dissolution was slow at the beginning (0-7 hr), rapid from 7 to 9 hr, and slow again at the end (9-20 hr). Complete dissolution was achieved in 20 hr, considerably longer than for the other actinide oxides previously tested. A point of interest is that in a similar experiment using only 4N HNO₃-0.1M ceric ammonium nitrate and ThO₂ (no filter media), the amount dissolved was only 25% after 20 hr. The reason for this difference is that the F⁻ catalyst is present in the filter media. Therefore, it is concluded that although

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the dissolution time was much longer for the ThO_2 , it is still an acceptable leaching reagent for ThO_2 from filter media.

Several experiments were completed using PuO_2 contaminated filter media from spent glovebox filters. These tests were made in order to determine how much $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$ is required for dissolution of PuO_2 from such filters. In one experiment, a 9.5-g sample of filter media was processed through a six-stage dissolution process. In the initial five stages, 4N HNO_3 -0.23M $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$ was used and in the sixth stage 12N HNO_3 -0.1N HF was used. The amount of $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$ used in each stage was 60 times the initial moles of PuO_2 present in the filter media. The overall decontamination factor obtained for the six-stage process was 833. In a second experiment, a 25.6-g sample of filter media was processed through a four-stage dissolution procedure. The leaching solution was 4N HNO_3 -0.30M $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$. The amount of ceric ammonium nitrate used in each stage was 120 times the initial moles of PuO_2 present in the filter media. The overall decontamination factor achieved in the four-stage process was 55.8.

The amount of ceric ammonium nitrate required cannot be based exclusively upon the amount of actinide present in the filter media. The reason for this is that other reactants such as pack-to-frame sealant* and other organic materials are present in the spent filter media. A high percentage of sealant was present in the 25.6-g sample and this is perhaps why less plutonium was dissolved. In other words, even though more ceric ammonium nitrate was used per mole of plutonium present, a lower percentage of the plutonium dissolved because a large percentage of the Ce^{+4} ions was depleted by reaction with the organics present. A formula for calculating the $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$ required per gram of filter media has not been developed. More studies must be done before the relationship can be determined.

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*This is a fire retardant polyurethane and a rubber base adhesive.

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DISCUSSION

BURCHSTED: After hearing your presentation, I wonder if a more appropriate title would be, "Product Recovery from Used HEPA Filters." Certainly, such a title would make the subject matter more accessible after the Proceedings have been indexed in other publications.

LEWIS: Thank you for that suggestion.

GUEVARA: I second the suggestion made by Burchsted. ;If decontamination were the appropriate topic, we would have liked to have heard more about economics and the ultimate disposal that is intended for the waste.

LOO: Would you describe the condition of the filter medium after the leeching process is completed?

LEWIS: After leeching, the filter medium is a white powdery substance.

LOO: Is it still intact?

LEWIS: No, the individual pieces have disintegrated and it has become a fine white powder.

DORMAN: Thank you, Mr. Lewis. It must have been quite a monumental experimental task to get all those results.

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CLOSING REMARKS OF SESSION CHAIRMEN:

GILBERT: In our first paper, Mr. McCormack reviewed a number of filters with different design and different media, some with prefilters, some without. With sodium oxide and hydroxide aerosols, he found considerable plugging, up to 25 in.w.g. I might advise him that filter media are only tested up to 20 in.w.g. Along the same lines, Dr. Hinds told us about the use of packed beds with different packings to be used with sodium aerosols. They exhibited considerable removal efficiency except for particles in the 0.5-1.1 μm range. In his brief report of the Government-Industry Meeting on Filters, Media, and Media Testing, Mr. Anderson did not have enough time to tell you that Johns-Manville described their latest quality control methods for measuring glass fiber diameters. Accurate measurement of fiber diameters is very critical for making filter paper that will meet nuclear grade filter criteria. They described their new program for making air permeability measurements that can be related to fiber diameter but they will continue to use the older Williams Freeness test simultaneously until all the users are familiar with the newer designations. Pursuing a DOE objective of reducing the volume of solid waste from spent filters that must be placed in storage or long-term burial, Mr. Woodward from Rocky Flats described their efforts to extend filter life by filter redesign, particularly filters subjected to chemical applications. In this endeavor, they are collaborating with the Bergman project at the Lawrence Livermore Laboratory.

DORMAN: I would also like to comment on the paper by Dr. Bergman because in 1960 I was sent Havlicek's original paper (most of it was written in his own fair hand) to referee for submission to the Journal. I have seen only about two references to that paper in the eighteen years that have elapsed since then. I think there was something by George Fielding a year or so ago and there has been an odd passing reference, but it never seemed to have been followed up very seriously. On the point of electric type filters, I would like to say there is another filter which is now being made in Europe and Holland based on the electric principle. Sheets of plastic material are subjected to voltages and the sheet is then stretched and fibrillated. This procedure produces quite fine fibers that can be fabricated into filters for air conditioning service or for dust respirators. They are quite efficient and have low resistance. However, they suffer, like most electric type filters, from the effects of oil which either dissolves the outer layers of the fibers or forms a Faraday cage that stops the electric effects. In addition, electric type fibrous filters tend to decay in efficiency with increase in loading of dust. For instance, one of the reasons why we did not use DOP in England in the old days when resin-wool filters were in vogue was that oil wrecked the wool resin filter in about 3 or 4 minutes. DOP increase filter penetration from 0.001% to 5 or 10%. Now, we find that even sodium chloride degrades the efficiency of electrified media. This means that electric type fibrous filters are always used. The papers by Dr. First and by Dr. Rivers were concerned with saving filter volume and they are, in a way, complementary to the work which is being done on dismantling filters. My own experience of what is termed the "European design filter," based on the LUWA filter, is that it's a satisfactory filter when it is used against normal dusts. I have had no experience with it in the nuclear industry, nor do I know how it will stand up against much rough usage. But, I shall be very interested to keep in touch with the work at AAF and Harvard and to hear what happens. On the paper by John Collins, I think there are arguments both for and against what he proposes. And, I think I would be very wise, as a non-United States chairman, to keep out of your internal arguments because I shall get clobbered by one side or the other and I want to go home. The last paper, presented by Mr. Lewis, obviously represented a very comprehensive series of tests and I can only admire the detail that he has presented. Obviously, he and his collaborators have spent a long, long time at it and produced a very good paper.