
CHAPTER 1

HISTORY OF THE DEVELOPMENT OF AIR CLEANING TECHNOLOGY IN THE NUCLEAR INDUSTRY

1.1 Brief History of Nuclear Aerosol Filtration

1.1.1 Early High-Efficiency Filter Paper Development for Military Gas Mask Use

In the early days of World War II, the British sent filter paper extracted from captured German gas mask canisters to the U.S. Army Chemical Warfare Service Laboratories (CWS) in Edgewood, Maryland.¹ The German filter paper was made of fine asbestos dispersed in esparto grass and had unusually high particle retention characteristics, acceptable resistance to airflow, good dust storage, and resistance to plugging from oil-type screening smokes (a deficiency of the resin-wool filters then used by the British forces). The CWS and the U.S. Naval Research Laboratory (NRL) reproduced the German-designed filter paper and had it manufactured in large quantities on conventional papermaking machinery by the Hollingsworth and Vose Company in Massachusetts. The first successful paper produced for the U.S. Navy contained Bolivian crocidolite and was called H-60. The paper produced for the U.S. Army also contained Bolivian crocidolite and was first designated H-64, but later renamed CWS Type 6. It was formulated from northern spruce sulfite and sulfate pulp (approximately 76 percent), cotton waste (approximately 15 percent), and Bolivian Blue crocidolite asbestos (approximately 14 percent). Penetration was 0.025-0.04 percent based on a methylene blue stain-intensity test procedure.²

The National Defense Research Council (NDRC), acting for the Armed Services, solicited the assistance of a number of university and industrial scientists in the search for better smoke filters. This effort resulted in important U.S. advances in the theory and technology of aerosol filtration. Up to this time, aerosol filtration theory had developed almost exclusively as an offshoot of water filtration knowledge. To meet then-current military requirements, however, researchers such as Nobel Laureate Irving Langmuir examined the physical basis for particle retention on fibers or small granules. Langmuir concluded that the principal mechanisms involved were: (1) interception, which affected suspended particles of sizes substantially greater than 1.0 micrometer (μm) in diameter when moving through a devious flow path in a bed of porous material; and (2) diffusion, which affected suspended particles with diameters substantially smaller than 1.0 μm .³ His analysis, later modified by Ramskill and Anderson⁴ to include inertia, indicated that the combined effects of these forces on a particle would be minimal when the particle was 0.3 μm in diameter. Langmuir advised testing gas mask filters with smoke of this particle size to determine their minimum retention efficiency and indicated that, when particles with diameters greater or smaller than 0.3 μm were present during field use of the gas mask, they would be removed at higher efficiencies than the test particles.

After the war, Victor LaMer⁵ of Columbia University performed many experiments to further examine Langmuir's theory of a minimum filterable particle size, concluding that efficiency declined as particle size decreased below 0.3 μm . Other research results confirmed a minimum filterable particle size, but not necessarily a diameter of 0.3 μm . This is understandable, as subsequent studies showed that forces not taken into account by Langmuir (particle inertia, flow rate, naturally occurring electrostatic charges on particles and filter media) can also affect collection efficiency. However history may judge the accuracy of Langmuir's theory, it profoundly affected U.S. filter technology and directly led to LaMer and Sinclair's development of the filter test used by the NDRC from 1942 through 1945. This filter test became the standard U.S. method for rating ultra-high-efficiency (i.e., absolute) filters.⁶ Before this standard, the U.S. Army Chemical Corps had

been using a test aerosol generated from methylene blue dye (dispersed from a water solution and dried). In 1963, W. H. Walton⁷ developed a sodium flame test to speed up testing of gas mask canisters because of the relative slowness of the methylene blue test procedure. This sodium flame test became the basis for the British standard test for high-efficiency filters.^{8,9}

1.1.2 Development of the High-Efficiency Particulate Air (HEPA) Filter

Protection against chemical warfare agents is required for operational headquarters, where wearing of an individual gas mask is impractical. To address this type of problem, the U.S. Army Chemical Corps developed a mechanical blower and air purifier known as a “collective protector” filter unit. Because relatively large air volume flow rates are required for effective use, the gas mask canister smoke filter (which uses CWS Type 6 filter paper) was refabricated into a filter constructed of deep pleats separated by a spacer panel and sealed into a rigid rectangular frame using rubber cement. The spaces between the teeth of the comb-shaped separators provided air passages to the depths of the pleats and were inserted front and back in alternate folds to direct contaminated air in and clean air out. The collective protector units were designed for use at the particulate removal stage by a combined chemical, biological, and radiological purification unit of the U.S. Armed Services. This development was highly fortunate, as later activities associated with the Manhattan Project created potential air pollution problems that could be solved only by using air filters with characteristics similar to those of the CWS filter. The U.S. Army Chemical Corps became the sole supplier of high-performance filters to the Manhattan Project, and later to the U.S. Atomic Energy Commission (AEC). In the late 1940s, the AEC adopted this type of filter to confine airborne radioactive particles in the exhaust ventilation systems of experimental reactors, as well as for most other areas of nuclear research. In this application, they were known as AEC filters or simply nuclear filters.

In recognition of their unusually high retention efficiency for very small particles, the U.S. Army Chemical Corps collective protector filters were also known as absolute, super-interception, and super-efficiency filters. The most widely used name, however, was HEPA filters, an acronym coined by Humphrey Gilbert, a former Manhattan Project safety engineer, from the title of a 1961 AEC report called *High-Efficiency Particulate Air Filter Units, Inspection, Handling, Installation*.¹⁰ A HEPA filter was defined as a throwaway, extended-medium, dry-type filter with: (1) a minimum particle removal efficiency of 99.95 percent (later raised to 99.97 percent) for a 0.3- μm monodisperse particle cloud; (2) a maximum resistance (when clean) of 1 inches water gauge (in.wg) when operated at rated airflow capacity; and (3) a rigid frame [now called “casing” in American Society of Mechanical Engineers (ASME) AG-1, *Code On Nuclear Air and Gas Treatment*].¹¹ extending the full depth of the medium (see **Figure 1.1**). HEPA filters have proven to be extraordinarily effective, reliable, and economical devices for removing radioactive and nonradioactive submicrometer-sized particles at a high rate of collection efficiency.

1.1.3 Early Nuclear Filter Developments in the United States

The U.S. Government was disturbed by the fact that components of the filter medium used in the CWS filters [Bolivian or African crocidolite (Blue Bolivian asbestos) and African esparto grass] had to be imported and could be difficult to obtain. After a variety of domestic cellulose fibers (yucca, Kraft, viscose) were used successfully by the NRL and the Hollingsworth and Vose Company as a replacement for esparto in trial runs, the AEC contracted Arthur D. Little, Inc. to develop a paper with equal or better filtration performance characteristics that could be manufactured entirely from fibers obtainable on the North American continent. Their investigations led them to examine coarse glass fibers as a substitute for cellulose, Canadian asbestos as a substitute for Bolivian Blue, and resin-stiffened, corrugated Kraft paper separators as a substitute for the comb-like separators in the CWS filter that had proved to be a significant obstruction to airflow.¹² The search for domestic sources of filter materials concluded successfully in 1951 with the development (partly sponsored by the NRL) of an all-glass-fiber paper made partly from super-fine glass fibers with diameters substantially less than 1.0 μm . As the domestic industry was able to produce unlimited quantities of glass

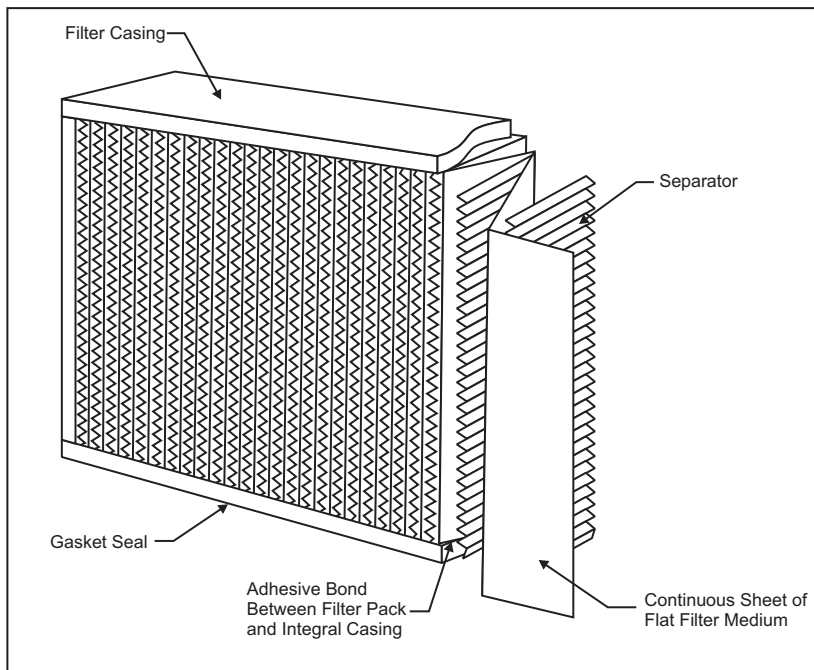


Figure 1.1 – HEPA Filter Design

fibers as small as 0.25 μm , asbestos was no longer needed. Abandonment of asbestos, which is difficult to disperse, allowed much greater control of manufacturing procedures and production of better, more uniform papers.

Because inclusion of some asbestos fibers in absolute (HEPA) filter papers containing glass fiber increases resistance to hydrogen fluoride and results in a slight cost reduction, some use of asbestos continued for a number of years after it was known that the papers could be made without asbestos. International concern about the toxic properties of inhaled asbestos fibers ultimately resulted in total abandonment of the commercial use of asbestos-containing filter papers, as well as the use of corrugated asbestos paper for

separators. Other materials were found that provided both improved resistance to chemical attack and fire resistance. Fires at the AEC's Rocky Flats Plant and in the Windscale graphite-moderated, air-cooled reactor in 1957 revealed the need for noncombustible effluent filters. The ability to make all-glass-fiber paper was a step in the right direction; but the separators, frames, and rubber cement used to seal the filter packs into the frames were all combustible. To overcome this problem, Arthur D. Little, Inc. was asked to develop a noncombustible absolute filter.¹³ They designed a prototype filter constructed from the glass-fiber filter paper prepared by NRL, corrugated asbestos paper separators stiffened by a water glass treatment, a perforated steel frame, and a refractory furnace cement for sealing the filter pack to the steel frame. The filter was completely fire-resistant, but it was heavy and the refractory furnace cement adhesive made the filter paper brittle, produced air leaks, and created a distressing tendency for the filter pack to separate from the steel frame. This filter assembly became obsolete after high chlorine- or bromine-content, self-extinguishing, flexible organic adhesives were introduced and Arthur D. Little, Inc. developed a fiber blanket seal that was compressed between the filter pack and metal frame.

The Hurlbut Paper Company and the Hollingsworth and Vose Company produced an air filter paper in the mid-1950s that was made from Fiberfrax fibers produced by the Carborundum Corporation. The Fiberfrax fibers were comprised of silicon oxide-aluminum hydroxide and could withstand temperatures up to 2,000 degrees Fahrenheit for long periods and in excess of 3,000 degrees Fahrenheit for shorter periods. Using this filter paper combined with loose Fiberfrax fibers of various grades, Flanders Filters, Inc., fabricated an all-ceramic filter (i.e., Fiberfrax paper, separators, filter-frame, and sealant) that was capable of performing satisfactorily at temperatures in excess of 2,000 degrees Fahrenheit and had extraordinary resistance to heat shock.¹⁴ However, it proved impossible to produce Fiberfrax fibers fine enough to provide filter efficiencies equal to those available with all-glass-fiber papers, and interest in Fiberfrax filters waned.

1.1.4 Commercial Development

After development of the absolute filter by Arthur D. Little Company, a manufacturing capability was installed at the Army Chemical Center in Edgewood, Maryland. Arthur D. Little also started the first commercial filter manufacturing company, the Cambridge Filter Company, which they sold shortly thereafter when they decided to restrict their efforts to research.

By 1957, three firms were fabricating absolute filters. Following allegations that defective filters were being delivered to its facilities, the AEC requested that sample filters from each of the three filter manufacturers be removed from AEC facility stocks and sent to Edgewood for inspection and testing. Seven of the 12 filters received by Edgewood had obvious defects upon removal from their shipping cartons.¹⁵ AEC facilities were advised to open and inspect the filters held in their stocks, and facility responses indicated a similar proportion of defects.

Based on these findings, the AEC initiated quality assurance (QA) inspection and testing of filter deliveries; installation of a test facility at Richland, Washington; and an agreement for QA testing by Edgewood for the eastern half of the United States. Oak Ridge, Tennessee, replaced Edgewood after installation of testing equipment there in 1964. A QA facility was activated at the Rocky Flats Plant in Golden, Colorado, in 1970. Facilities at both the Rocky Flats and Richland sites have been decommissioned and dismantled.

A Government-Industry Filter Committee was established at about this same time with voluntary participation from representatives of filter manufacturers, filter medium makers, the sole supplier of glass fibers, users, and Government agencies and organizations, including the Army Chemical Center and the NRL. Discussion sessions were held before the biennial AEC Air Cleaning Conferences, and working sessions were convened at the Underwriters Laboratories (UL) in Chicago, Illinois. Topics ranged from the aging of glass fibers to the integrity of shipping cartons. The Committee provided guidance to the Army Chemical Center concerning military standards for fire-resistant filters and its glass fiber filter medium, and also advised UL in establishing their UL-586 standard for filter heat resistance.¹⁶ The Committee was also responsible for considerable technology exchange (in view of the relative newness of the glass fiber filter medium and the undeveloped technology for its fabrication into filters).

1.1.5 Development of HEPA Filter Standards

With the Army's issue of Military Specifications MIL-F-51068, *Filter, Particulates, High-Efficiency, Fire-Resistant*,¹⁷ for the fire-resistant filter and MIL-F-51079, *Filter Medium, Fire-Resistant, High-Efficiency*,¹⁸ for the glass fiber medium in the early 1960s, Edgewood abandoned its manufacture of the cellulose-asbestos filter and turned to commercial procurement. These standards documents remained in service until 1994, when due to changing requirements, the availability of new materials, improved instrumentation, advanced technology, and a U.S. Department of Defense emphasis on consensus standards, the U.S. Army announced it would no longer maintain MIL-F-51068 and MIL-F-51079 in active status. Both of these military standards were incorporated into ASME AG-1,¹¹ Section FC, which is administered by the Committee on Nuclear Air and Gas Treatment (CONAGT). Improvements were incorporated into the standards with the concurrence of the other military services and the U.S. Nuclear Regulatory Commission (NRC).

The HEPA filter design used by the U.S. nuclear industry is nearly identical to the one used in the United Kingdom and has been the mainstay of the nuclear industry for the past 5 decades. Additional progress was made in documenting and codifying standards for filter installation and testing with the AEC's issuance of the original Regulatory Guide 1.52, *Design, Testing and Maintenance Criteria for Engineered-Safety-Feature Atmospheric Cleanup System Air Filtration and Adsorption Units of Light-Water-Cooled Reactors*,¹⁹ in 1973. [Unknown to most, Mr. Humphrey Gilbert, mentioned in Section 1.1.2, and Dr. Roger Zavadoski were the primary authors of

this Regulatory Guide.] Further progress was made with the American National Standards Institute's issuance of ANSI N509, *Nuclear Power Plant Air Cleaning Units and Components*,²⁰ and ANSI N510, *Testing of Nuclear Air Cleaning Systems*.²¹ Although these two standards were intended to apply only to the construction and testing of engineered safety systems in U.S. civilian nuclear power plants, the major part of each standard can be and often has been applied with salutary results to air cleaning systems in all manner of nuclear facilities in the United States (including DOE facilities) and abroad. CONAGT has transferred many sections of the former ASME N509²² and N510²³ into ASME AG-1.¹¹ The contents of the early editions of these two standards were substantially incorporated into NRC Regulatory Guide 1.52, Revision 1, *Design, Testing and Maintenance Criteria for Engineered Safety Feature Atmospheric Cleanup System Air Filtration and Adsorption Units of Light-Water-Cooled Nuclear Power Plants*.²⁴ Some standard-setting agencies in other countries with a significant nuclear power establishment have prepared and issued similar standards that differ only in details. The principal modification to the military standards since 1968 focused on requirements for filter medium resistance to radiation (for prolonged filter effectiveness following a core-disruptive accident). For procurement ease, the military service (Edgewood Arsenal) qualified HEPA filter paper and assembled filters manufactured by a number of producers and published their names in a Qualified Products List (QPL).²⁵

Table 1.1 lists important developments relating to filtration and the year of development.

Table 1.1 – Summary of Important Dates for Nuclear Air Cleaning Filtration

Year	Publications/Actions
1950	MIL-STD-282, <i>Filter Units, Protective Clothing Gas-Mask Components and Related Products: Performance Test Methods</i>
1950	Stack Gas Committee
1950s	Arthur D. Little Co., <i>Fire Resistant Media</i>
1957	Air Cleaning Conference, "Filter Quality Problems"
1959	Air Cleaning Conference, "Filters Sent to Edgewood"
1959	Government/Industry Safety Committee
1959	UL-586, <i>High Efficiency, Particulate, Air Filter Units</i>
1961	High Efficiency Particulate Air Filter Units, TID 7023, Gilbert and Palmer
1962	Hanford (AEC/DOE) Filter Test Facility
1962	MIL-F-51068, <i>Filter, Particulates, High-Efficiency, Fire-Resistant</i>
1963	MIL-F-51079, <i>Filter Medium, Fire-Resistant, High-Efficiency</i>
1963	Flanders Inc. - Filter Media Production
1966	ORNL/NSIC-13, <i>Filters, Sorbets and Air Cleaning Systems as Engineered Safeguards in Nuclear Installations (Nuclear Air Cleaning Handbook, 1st Edition)</i>
1968	AACC CS-IT HEPA FILTER (IES), <i>Tentative Standard for HEPA Filters</i>
1968	ASHRAE 52.68, <i>Method of Testing Air Cleaning Devices Used in General Ventilation for Removing Particulate Matter</i>
1969	ORNL/NSIC-65, <i>Nuclear Air Cleaning Handbook, 2nd Edition</i>
1971	ANSI N-45.2, <i>Requirements for Quality Assurance Programs for Nuclear Power Plants</i>
1971	ANSI N-45.8 CONHET
1972	Flanders Inc. - Manufactures Glass F-700 Media
1973	REGULATORY GUIDE 1.52, <i>Design, Testing and Maintenance Criteria for Engineered Safety Feature Atmospheric Cleanup System Air Filtration and Adsorption Units of Light-Water-Cooled Nuclear Power Plants</i>
1973	REGULATORY GUIDE 3.12, <i>General Design Guide for Ventilation Systems of Plutonium Processing and Fuel Fabrication Plants</i>
1975	ANSI N510, <i>Testing of Nuclear Air Treatment Systems</i>
1976	ASME CONHET
1976	ANSI/ASME N509, <i>Nuclear Power Plant Air Cleaning Units</i>
1976	ERDA 76-21, <i>Nuclear Air Cleaning Handbook, 3rd Edition</i>
1978	Flanders Inc. - Manufactures Last Glass/Asbestos Media

Year	Publications/Actions
1979	REGULATORY GUIDE 1.140, <i>Design, Inspection and Testing Criteria Air Filtration and Adsorption Units of Normal Atmosphere Cleanup Systems in Light-Water-Cooled Nuclear Power Plants</i>
1978	NE F3-41T, <i>In-Place Testing of HEPA Filter Systems by Single-Particle, Particle-Size Spectrometer Method</i>
1980	Flanders Inc. – Manufactures Last Asbestos Separators
1984	DOE HEPA FILTER/TEST STANDARDS NE F3-43, -44, -45, Nuclear Standards, <i>Nuclear Standard Quality Assurance Testing of HEPA Filters, DOE Filter Test Facilities Quality Program Plan, and Specifications for HEPA Filters Used by DOE Contractors</i>
1984	NE F3-42, Nuclear Standard, <i>Operating Policy of DOE Filter Test Program</i>
1984	ASME AG-1, <i>Code on Nuclear Air and Gas Treatment</i> , 1st Edition
1993	ASTM-F-1471-93, <i>Standard Test Method for Air Cleaning Performance for HEPA Filter Systems</i>
1997	DOE-STD-3020-97, Replaced NE F 3-45 HEPA Filter Standard, <i>Specification for HEPA Filters Used by DOE Contractors</i>
1999	ASHRAE 52.2, <i>Method of Testing General Ventilation Air Cleaning Devices for Removal Efficiency by Particle Size</i>
2003	ASME AG-1, <i>Code on Nuclear Air and Gas Treatment</i> , Update
2003	<i>Nuclear Air Cleaning Handbook</i> , 4th Edition

1.1.6 Further Development of the HEPA Filter

Thin, corrugated aluminum-alloy separators completely replaced asbestos, thermoplastics, and resin-treated Kraft paper to assure fire-resistance. Stainless steel is often selected because of its resistance to severe chemical attack, but aluminum-coated plastic is satisfactory for less corrosive service. Improved resistance to wetting, an issue of major importance for engineered safety system filters in water-cooled reactors, was developed by applying water-repellent chemicals to the filter paper. For such applications, it has become standard practice to install the filters with the paper folds in the vertical position so that any water droplets captured on the surface of the paper will drain to the bottom of the filter.

1.1.7 Introduction of HEPA Filters for Treating Reactor Effluent Gases

The first nuclear reactor fitted with effluent high-efficiency air filters is believed to have been the graphite-moderated, air-cooled unit at Oak Ridge National Laboratory (ORNL) in Oak Ridge, Tennessee. The initiating event was the discovery in 1948 of radioactive particles up to 600 μm in size on the ground around the reactor stack. A reinforced concrete filter house capable of handling 140,000 cubic feet per minute (cfm) of air at a temperature of 215 degrees Fahrenheit and a negative pressure of 50 inches water gauge (in.wg) was constructed to prevent further emissions.²⁶ This was also one of the first installations to use prefilters to extend the life of absolute filters as a means of reducing air cleaning costs. The filtration system contained 1-inch-deep resin-bonded fiberglass prefilters that removed the coarsest dust fraction, followed by 24 inches \times 24 inches \times 11 1/2 inches Army Chemical Corps cellulose-asbestos (later designated AEC No. 1) units in plywood frames. Design efficiency was 99.9 percent for particles down to 0.1 μm . The high-efficiency filters were changed when airflow resistance increased from 1 in.wg to 5 in.wg over a period of about 2 1/2 years. It was found that the service life of the absolute filters could be extended to more than 2 years by changing the prefilters two to three times per year. Although there have been situations where a cost analysis has failed to show an advantage from using prefilters, most installations seem to benefit from using cheaper prefilters. Interest in the use of metal prefilters continues because, in addition to coarse particle filtration, they provide fire and blast protection by acting as baffles and fire screens.

1.1.8 HEPA Filter Quality Assurance

During the 1960s, major efforts in the United States were directed toward standardizing manufacturing and test criteria for paper and fabricated filters, with special emphasis on fire and water resistance. Manufacturer testing of each individual filter for collection efficiency and airflow resistance has always been a unique requirement for filters intended for use in nuclear service. The results of each test are noted on the filter frame to ensure the filter meets the requirements of applicable standards. Initially, the efficiency standard for 0.3- μm test aerosols was 99.95 percent, but it was raised to 99.97 percent after commercial filter manufacturers found ways of improving their materials and assembly techniques to a degree that enabled them to turn out filters exceeding the required particle retention efficiency by more than an order of magnitude. These new filters also featured improved resistance to corrosive chemicals, fire, and radiation.

Similar filter efficiency standards were developed in Great Britain (using a nebulized salt aerosol),⁸ France (using a nebulized uranine aerosol),²⁷ and Germany (using a paraffin oil aerosol).²⁸ Because of differences in measuring filter efficiencies, considerable effort has been expended (with indifferent success) on laboratory studies to develop conversion factors that would translate the filter efficiency measurements made by one method to equivalent values derived using a different measurement method.²⁹ It would be convenient if everyone used the same filter test method, but this is unlikely in the foreseeable future.

The wide diversity of aerosols generated in the nuclear industry raises an important question regarding the relevance of the qualification test procedures utilized. For example, the aerosols predicted to be present inside the confinement vessel of a power reactor following a loss-of-coolant accident (LOCA) are certain to be very different from the test aerosols and efficiencies observed during the standardized qualification tests and are not necessarily the results that will be obtained in practice. They may be better or worse, depending on the characteristics of the aerosol challenge. However, passing a standardized qualification test gives reasonable assurance that the filters have been produced from high-quality components and carefully assembled to exacting standards. Therefore, the standard qualification test results should be viewed as an index of merit (an indication of quality) rather than a quantitative description of filter efficiency under unknown or ill-defined operating conditions.

Nevertheless, about 1990, the U.S. Department of Energy (DOE) undertook a program designed to define HEPA filter efficiency more precisely. This involved the use of an intercavity laser particle size spectrometer capable of counting and sizing aerosol particles down to approximately 0.08 μm under careful laboratory manipulation. The impetus for this program was the discovery that the monodisperse 0.3- μm test aerosol (when defined using methods developed during the 1940s) was neither monodisperse nor always 0.3 μm .²⁹ Filter efficiency studies conducted at Los Alamos National Laboratory produced the following results:

- The most penetrating particle size for all-glass-paper HEPA filters operated at the design airflow rate is close to 0.1 μm .
- A new HEPA filter acceptance standard was developed that used a polydisperse aerosol, but this method counted only 0.1- μm particles upstream and downstream of the filter to rate particle retention efficiency.³⁰
- Programs were conducted at DOE filter test stations to improve the characteristics of the aerosol used for routine filter testing (e.g., making the test aerosol more uniform in size and closer to an average size of 0.3 μm).

To a significant degree, the establishment of AEC QA filter test stations in 1960 made it imperative for filter manufacturers to institute their own rigid quality control practices to avoid product rejection. For example, 49 percent of filters manufactured prior to 1960 were rejected, whereas only 5 percent were rejected during the following 8 years.³⁰ By 1978, the rejection rate had declined to a point where the NRC was willing to

forego QA filter test station inspection of filters intended for use in engineered safety feature (ESF) systems in commercial nuclear power plants. The basis for this decision was that the marginal increase in the reliability of tested filters no longer justified the additional cost. It should be noted that commercial ESF system filters are usually in a standby mode, in a clean system, and assigned minimal removal efficiencies (relative to DOE facilities) in their safety basis. DOE continues to require the use of a filter test station because contaminated processes continuously challenge DOE filter systems.

Only one DOE QA filter test station, now called the Filter Test Facility (FTF), is currently operating. The rejection rate of filters tested there has been as high as 18.7 percent and as low as 1.6 percent in recent years. The rejection rate continues to fluctuate, indicating that the FTF is still necessary. The Secretary of Energy mandated continued use of the FTF in 2001.

Considering the large number of specifications, requirements, and standards that have been proposed and adopted for HEPA filters, it is clear they are among the most extensively and thoroughly documented devices in the entire air filtration spectrum.

1.1.9 HEPA Filter Application Assurance

In spite of the many improvements in absolute (HEPA) filters, it was discovered as early as the initial installation of HEPA filters at the ORNL graphite reactor that the full capabilities of improved filter performance were not always achieved due to damage during shipment or faulty installation. Consequently, it has become routine to conduct in-place testing of all filter installations using methods initiated and developed at ORNL prior to startup of new facilities and periodically thereafter. A great deal has been learned about the correct design of filter housings and filter installation methods from in-place testing. For example, considerable difficulty was experienced in conducting tests at old installations due to lack of easy access to the filter structures. It became clear that suitable facilities for in-place filter testing must be designed into all new systems as part of the construction specifications.

The value and effectiveness of correctly designed and installed nuclear-grade aerosol filtration systems are illustrated by the very different events that took place at the Three Mile Island-2 (TMI-2) and Chernobyl reactors. During the March 1979 LOCA at TMI-2, two 30,000 CFM filter systems prevented essentially all of the particulate material and the bulk of the radioiodine released to the Auxiliary Building from being released to the environment.³¹ Consequently, release of radioactive particles to the environment was negligible. The outcome was very different, however, during the April 1986 fire at Chernobyl Unit 4, where engineered safeguards did not include complete confinement with air filtration systems. The widespread apprehension caused by that accident is likely to produce a demand for still higher collection efficiency and greater filter resistance to internal disruptive events (fires, explosions) and to external natural disasters (earthquakes, tornadoes). Germany³² and the United States³² have responded to this by developing filters composed of stainless steel fibers.

1.1.10 Increasing Airflow Capacity of HEPA Filters

Although British filter construction methods and materials closely paralleled American ones, manufacturers in other European countries developed a different HEPA filter design that is now produced by some U.S. manufacturers. Instead of filter paper pleats that extend the full depth of the filter frame, the paper is folded into mini-pleats about 20-millimeters (mm) deep with a pitch of 3 mm. Adjacent pleats are separated by ribbons or threads of glass, foam, or plastic. A full-size filter is assembled from several component panels of this construction and arranged around a series of V-shaped air passages. This design allows considerably more filter paper to be incorporated into a given volume, making it possible to replace a standard 24 inch \times 24 inch \times 11 1/2 inch U.S. filter unit with one of identical dimensions that: (1) can handle volumes

up to 1,900 CFM instead of 1,000 CFM at a clean filter resistance of 1 in.wg, and (2) can meet the maximum test aerosol penetration standard of 0.03 percent at the higher volumetric flow rate.

A U.S. manufacturer has fabricated a different filter that does not use separators. The corrugations are made by vacuum-molding the wet filter paper onto narrow longitudinal ridges while it is still on the paper-making machine, then accordion-pleating the paper as it comes off the machine.³³ The preformed corrugations are impressed into the paper at a slight angle to the run of the sheet so that, when folded, the pleats in alternate layers resist nesting. A later development of this process is to impress dimples into the forming paper so that, when folded, the dimples prevent alternate paper layers from touching each other. This filter construction method is different from the one used for the older mini-pleat filters in that the filter pack is mounted into the filter frame in the usual way (i.e., perpendicular to the direction of airflow) rather than as a number of 20-mm-deep panels arranged inside the filter frame in a series of V-formations. A 6-inch-deep mini-pleat filter without separators contains the same area of filter paper as the 12-inch-deep separator type. This filter has been placed into service, but there is no experience to report concerning nuclear applications.

1.1.11 Disposal of Spent Filters

It costs more to dispose of a contaminated spent filter than its initial purchase price, which reflects the difficulties associated with handling contaminated wastes and the shrinking number of authorized disposal sites. During the early years of the nuclear age (when HEPA filters were constructed with wooden frames, corrugated separators, heavy Kraft paper, cellulose-containing filter paper, and conventional rubber cement), high-temperature incineration resulted in a 99 percent reduction in bulk. At the time, this was considered the best way to handle used filters, and a number of incinerators were constructed and used to reduce the bulk of all combustible contaminated wastes, including spent filters. However, the incinerators quickly became contaminated and proved difficult to safely operate and repair. To protect the environment, HEPA filters were installed as the final flue gas cleaning element, but they proved to have a short life in incinerator service. In addition, processing the spent flue gas filters through the same incinerator they were installed to serve greatly increased the burden on the incinerator, thereby reducing productive throughput and elevating costs.

During the 1960s, as a result of the introduction of noncombustible elements into the structure of HEPA filters intended for nuclear service and the introduction of heavy presses designed to crush HEPA filters into a small volume for ground burial at little cost, outmoded high-temperature volume reduction incinerators were shut down and dismantled. Where recovery of transuranic elements from spent filters remained a requirement, devices were developed to extract only the filter paper from the frame for chemical or high-temperature treatment. The remainder of the filter was disposed of by crushing and burial.

The rapidly escalating cost of land disposal for radioactive wastes, in addition to new requirements for corrosion- and leak-proof containers that substantially increase the bulk of the waste package, have combined to renew interest in volume reduction of wood frame filters by high-temperature incineration in spite of an obvious incompatibility between the need for noncombustible filters and the need to minimize disposal costs via high-temperature volume reduction. Exclusive use of HEPA filters without separators help reduce the residue from incineration. When using metal frames and corrugated aluminum separators, alternatives include punching out the filter pack in a high-pressure press for volume reduction and decontaminating the metal parts via chemical treatment. Incineration of contaminated HEPA filters continues to present formidable operating difficulties and high costs. Additional difficulties are experienced when the substances collected on filters are classified as both hazardous chemical and radioactive wastes.

1.2 Deep-Bed Sand and Glass Fiber Filters

Although HEPA filters came to dominate aerosol confinement for most nuclear applications, from the beginning there were other filter innovations of note. When a high-activity level was detected at the Hanford,

Washington, site in 1948 and traced to radioactive particles emitted from the chemical processing ventilation stacks, the chemical engineering practice of using deep beds of graded granular coke to collect mists escaping from contact sulfuric acid plants was recalled, and a number of large sand filters were constructed during the late 1940s and early 1950s at both the Hanford and Savannah River Sites (SRS).³⁴ The sand filter construction closely followed the deep-bed (40- to 120-inch deep), graded-granule techniques for building granular filters that were widely accepted at sulfuric acid manufacturing plants and for the purification of municipal drinking water supplies. These filters had collection efficiencies for particles greater than 0.5 μm that compared favorably with the best fibrous filters then available. They operated at a superficial face velocity of 6 feet per minute, an initial pressure drop of 8 in.wg, and an activity reduction of 99.7 percent. Additional units were later built at SRS, and each has given many years of continuous service. Such deep-bed sand (DBS) filters offer several advantages. They offer a higher design airflow resistance and lower retention efficiency than may be obtained using absolute filters. They also are nonflammable and largely unaffected by condensed water and strong acids. In addition, they provide a substantial heat sink in the event of fire or explosion. Freedom from servicing and replacement over many years is another important advantage when the collected material is intensely radioactive. DBS filters are not completely maintenance-free, however. Collapsed laterals have led to replacement of tons of sand and increased surveillance. The disadvantages of DBS filters are that they are large, expensive to operate and build, and nondisposable.

Rapidly emerging glass fiber technology during the 1940s and 1950s shifted attention to the use of very deep beds (10 inches or more) of curly glass fibers in combination with HEPA-quality final filters as a satisfactory substitute for sand filters when treating gaseous effluents from chemical operations.³⁴ These proved to be more efficient and to have lower airflow resistance than the sand filters they replaced. Deep-bed glass fiber filters have been used at Hanford for several decades on the Purex process effluent stream, and a similar installation is in place at DOE's Idaho Chemical Plant. They also have withstood the buildup and mitigation of potentially explosive nitrates.

There has been interest in sand filters for emergency confinement venting for light water reactors. An installed Swedish confinement venting system known as FILTRA features large concrete silos filled with crushed rock. These silos were designed to condense and filter steam blown from the confinement and to retain at least 99.9 percent of the core inventory.³⁵ Later designs for confinement venting utilized wet systems to remove gaseous radioiodine.

1.3 Brief History of Gas Adsorption

[Note: The following discussion concerns adsorbents used to capture gaseous and volatile fission products and is included as history only. Adsorbents are commonly used for iodine removal in commercial nuclear power plants (see AG-1¹¹ for more information). Current DOE nuclear applications predominantly rely on HEPA filters rather than adsorbents. However, references to adsorbents will be found in nearly each chapter.]

1.3.1 Introduction

Iodine in its many chemical forms is probably among the most extensively studied fission products produced in the nuclear industry. The generation, release mechanism, properties, forms, trapping and retention behavior, and health effects of iodine-131 have been the subject of numerous studies, but a comprehensive understanding of the significance of its release to the environment and integration of the chemical technology into protection technology may remain incomplete in some aspects. The technology associated with the removal and retention of all iodine isotopes is similar to that for iodine-131, but interest in removal efficiency has shifted somewhat toward the importance of long-term retention with the increasing half-life of the iodine isotope.

A removal technology for the radioactive noble gases (krypton, xenon, and radon) using adsorbents also has been studied extensively. This removal technology has become a standard control method for boiling water reactor (BWR) offgas decontamination and has replaced pressurized tank retention for pressurized water reactor (PWR) offgas control. A similar technology can be used to hold up the relatively long-lived krypton-85 contained in reprocessing offgases.

Volatile metal compounds such as ruthenium and technetium can be removed from gas streams by adsorption, but a solid-surface-supported chemical reaction is often necessary for good retention. Removal technologies for carbon-14 and tritium also involve the use of adsorbents, either as collecting agents or as catalysts for conversion to other, more easily removed compounds.

Vapor recovery by adsorption was a well-established chemical engineering unit operation process prior to nuclear technology development for weapons and power production. Generally, vapor recovery systems utilized beds of activated carbon that were 24 inches deep or more and often consisted of two or more identical units in parallel, so that one could be onstream while a second was being desorbed by low-pressure steam and a possible third was undergoing cooling after steam desorption. These multi-bed arrangements enabled continuous operation of vapor production processes.

Adsorbents of various types, both impregnated and unimpregnated, became widely used during and following World War I (WWI) in military and civilian gas mask canisters and cassettes for removing a wide range of toxic substances from breathing air. Activated carbon derived from nut shells was used in the U.S. Army service gas mask during WWI. Later, the activated carbon used in the service gas mask was derived from coal and impregnated with metals that catalyze reactions with gas warfare agents. Activated carbon also was used to treat ventilation air in special applications such as removing sulfur dioxide and ozone from air supplied to libraries housing rare book collections to prevent paper embrittlement. Ventilation applications used shallow beds of activated carbon, generally 1 inch or less, because complete removal of outdoor contaminants was seldom a requirement and low airflow resistance was essential to prevent unacceptable fan noise levels. The theoretical basis for adsorption processes was greatly advanced by the need to develop gas mask applications during WWI, and Langmuir made an early theoretical analysis of physical adsorption. Thus, there was a considerable body of knowledge available on the application of adsorbents, especially for activated carbon, when the nuclear industry developed a need for this technology.

Control of iodine emissions from chemical processing of spent nuclear fuel was initially done by liquid scrubbing using caustic solutions, and sometimes with the addition of sulfate salts, but retention efficiency by scrubbing seldom exceeded 90 percent. To improve iodine retention efficiency for dissolver offgas cleaning, activated carbon beds were added to the caustic scrubber at DOE's Idaho Chemical Plant in 1958, where they were reported to provide additional decontamination factors of 10 to 30. Silver-plated Fiberfrax fibers also were investigated at the Idaho Test Station for use as a combined particulate filter and iodine retention device for hot calciner offgas cleaning. Other studies of this nature were conducted with silver-plated copper filaments, and an iodine decontamination factor of 10 was reported.

Iodine releases to the atmosphere in the event of a reactor accident became a major concern as the nuclear industry began its rapid expansion during the early 1960s, and attention focused on iodine removal during normal and abnormal conditions at ambient and elevated temperatures. An iodine decontamination factor of 10 was reported. At ORNL, studies were conducted on activated carbon beds for the holdup of radioactive fission gases generated during the operation of nuclear reactors and during nuclear fuel reprocessing. The principal area of interest was delaying release until short-half-life isotopes decayed to levels that were acceptable for release. This approach utilized conventional theoretical plate equations.

The application of adsorbents for noble gas retention was developed at ORNL. The concept involves self-regeneration of the adsorbent due to decay of the noble gases to solid daughter products as they pass through very deep adsorbent beds that require a long time for passage and results in the successive extinction of noble

gas radioisotopes (i.e., those with the shortest half-lives disappear first). This technology is generally used to decontaminate all noble gas isotopes (except krypton-85 because of its relatively long half-life—nearly 11 years). The process is particularly well suited to treat BWR offgas streams and was applied first at the KRB site in Germany. The first BWR installation in the United States was the Interim Offgas System at the Vermont Yankee Plant. It was succeeded by the Advanced Offgas System at the same site. Earlier technology involved ambient temperature systems. Cooled or refrigerated systems were later designed by the General Electric Company.

Storage tanks were used for PWR degasifier gas processing at first, but a continuous-flow adsorption system was installed at the Seabrook nuclear power plant, the first for a U.S. PWR. Design parameters for noble gas adsorption systems were established on a more systematic basis than was the case for control of radioiodine, and the few problems that have occurred with these plants were related to improper humidity control or accidental wetting of the carbon prior to operation. Two temperature excursions have been reported in these systems—one at ORNL, where an oxygen stream was being decontaminated, and one at the Brown's Ferry nuclear power plant, where a hydrogen recombiner malfunctioned.

Testing of Iodine Adsorbents

The current test protocol is ASTM D3803-89,³⁶ which superseded RDT M-16.³⁷ Both standards have numerous typographical and editorial mistakes, such as inaccurate decay constants for iodine-131 and inconsistencies in time duration between the text and tables. In addition, both are merely guides as far as equipment setup is concerned, but the critical parameters listed in both Table No. 1 and Section 13 of ASTM D3803-89³⁶ specify reporting requirements.

Testing of Noble Gas Adsorbents

The results of noble gas delay cannot be correlated because important test parameters either were not reported or were not standardized. Omissions include the unspecified moisture content of the adsorbent, relative humidity of the gas, and duration of pre-equilibration for the experiment. In some cases, tests involved only a few grains of carbon, and the results have been extrapolated to full-size systems with bad results.

Operating Experience with Iodine Adsorption

Several important lessons concerning iodine control were learned from the TMI-2 accident. The first is that conventional iodine release and transport theories were incorrect. Most of the iodine stayed in the liquid phase or plated out in the confinement vessel. The total amount of iodine that reached the operating filter adsorber trains can be conservatively estimated at 150 curies (Ci), of which approximately 15 to 32 Ci were released to the environment. This value, when compared with approximately 13×10^6 Ci of xenon-133 released (the approximately hundreds of millions of iodine curies released into the containment), is a good indication of a lack of the predicted partitioning of iodine species into the airstream. One indication of the iodine species distribution showed a predominance of methyl iodide, followed by elemental iodine. The system available for controlling iodine releases was comprised of two trains in the Unit 2 Auxiliary Building, identified as trains A and B, and two trains in the Fuel Handling Building, identified as trains A and B. The Auxiliary Building trains were not classified as ESFs. They captured approximately 12 to 14.6 Ci of iodine and released approximately 1.2 to 1.8 Ci. The Fuel Handling Building filters captured approximately 36 to 48 Ci of iodine and released approximately 5 to 15 Ci.

1.3.2 Radiochemical Processing

The quantity of radioiodine used in radioactive tracer studies is small compared to the concentrations present in power reactors, but the variety of radioiodine-containing organic compounds is greater. Based on available theoretical and experimental data, the removal efficiency of impregnated nuclear carbons for many organic compounds is lower than for methyl iodide. Furthermore, most radioiodine decontamination systems found in connection with laboratory fume hoods are inadequate even for methyl iodide, as they usually only contain a depth of 1 inch of some unimpregnated carbon that has not been specifically qualified for this intended use. For laboratory hood service, carbon depth should provide at least a 0.25- to 0.50-second residence time and should permit removal of representative samples for periodic laboratory testing to determine remaining service life. Representative samples should be removed at least every 720 hours of continuous use and should be tested under conditions corresponding to the hood effluent conditions with respect to relative humidity, temperature, and the presence of compounds that compete with the radioiodine species for adsorption sites. For example, when relative humidity is variable, the adsorbent should be tested at the maximum relative humidity conditions likely to be present to obtain conservative values.

1.3.3 Fuel Reprocessing Plants

The isotope of importance in the effluent gases from fuel reprocessing systems is iodine-129, which has a half-life of 1.7×10^7 years. Generation of gaseous iodine-129 occurs in the presence of oxidizing acid gases such as nitrogen oxide under very-high-humidity conditions, and often when there are high concentrations of competing organic compounds. This is a highly demanding environment for adsorption media. At the beginning, reprocessing effluent treatment in the United States usually involved liquid scrubbing with alkaline solutions. However, there are anecdotal reports of a packed bed scrubber at Hanford that utilized silver dollars for the packing to make the captured iodine more insoluble as silver iodide. Although alkali, mercuric nitrate, and hyperazeotropic nitric acid absorption systems are still used for this purpose, direct removal of iodine using solid adsorbents has been gaining favor in treating the gaseous effluent at newer fuel reprocessing plants. The use of solid adsorbents for this service was first evaluated at the SRS with activated carbon, but it proved to be unstable in the dissolver offgas environment. In 1968, a switch was made to a silver-impregnated inorganic adsorbent. The solid adsorbents under consideration included primary silver-containing materials such as silver-exchange zeolites and silver-impregnated adsorbents, where the adsorbent acts as a carrier for the silver-iodine chemical reaction. Due to the relatively high cost of silver, it is important that as much silver as possible is utilized before exhaustion of the adsorbent system. Numerous studies have been conducted to evaluate these materials for full reprocessing service.

The most commonly used adsorbents for dissolver offgas treatment include AC6 120, a silver-nitrate-impregnated, high-silica-base adsorbent; a silver-and-lead-nitrate-impregnated, high-temperature base adsorbent; and silver-exchange zeolites and mordenites. Several reaction mechanisms lead to various silver-iodine compounds. The most common compound for both elemental and organic iodine is silver iodide, which is very stable except in a high-temperature hydrogen environment where reduction to elemental forms occurs.

1.3.4 Power Reactors

The first major U.S. effort related to control of radioiodine from reactors consisted of design studies of confinement systems for the nuclear-powered commercial ship N.S. Savannah and the Hanford N Reactor.^{36, 37, 38} At that time, control of elemental iodine was of primary interest, mainly because data from various prior accidents failed to differentiate iodine forms. A process-engineering solution to iodine retention was proposed that recommended 12-inch-deep carbon beds operated at high velocity with a 0.5-sec residence time. In the United States, however, the heating, ventilating, and air-conditioning (HVAC) shallow-bed

model was adopted by the nuclear industry, and shallow beds of carbon became the predominant method for iodine capture.

The U.S. activated carbon adsorber design was based on a series of relatively short-term laboratory experiments using fresh carbon, clean carrier gas, and nonsystematic iodine inlet concentrations. Results indicated an iodine removal efficiency for 0.8- to 1.0-inch-deep carbon beds that could not be obtained in practice. Typically, the early installations were constructed in pleated form and contained 44 to 55 pounds of carbon for every 10,000 CFM of airflow. This design became known as a Type I Adsorber Unit. It was later found that, under high-humidity conditions (greater than 70 percent relative humidity), shallow carbon beds were incapable of high-efficiency removal of organic iodides, particularly methyl iodide. In addition, it was accidentally discovered that: (1) isotope exchange would take place on carbon surfaces, and (2) gas mask carbons impregnated with tertiary amines to control low-molecular-weight chemical warfare agents containing organic halides would also react with radioactive organic halides. This discovery led to the use of carbons impregnated with stable iodine or iodide salts to control methyl iodide by isotope exchange, as well as the use of amine-impregnated carbons to control methyl iodide by complex formation.

Although laboratory experiments with unimpregnated carbons indicated that a 1-inch bed performed acceptably for elemental iodine removal when the exposure was a short duration and the carbon was fresh, a minimum acceptable bed depth of 2 inches was needed under ideal conditions for the impregnated carbons used for methyl iodide removal. This led to development of a tray-type design for nuclear adsorber units consisting of two 2-inch-deep military-type adsorber trays that were attached to a 24-inch \times 24-inch face plate for mounting in ladder frames. This adsorber design became known as a Type II Adsorber Unit. It provided a 0.25-second gas residence time in the carbon and operated at a gas velocity of 40 feet per minute (fpm).

Standardization of the external dimensions of the tray-type units did not occur for many years, and there are currently approximately 10 different adsorber sizes in service in the United States. This creates logistical difficulties for warehousing spares and obtaining fast replacements in case of an accident. For example, between the two reactors on the TMI site at the time of the TMI-2 accident, there were four different adsorber shapes and sizes—three of them supplied by the same vendor.

In the beginning, criteria for the selection of adsorbent media were not well standardized in the United States. Based on short-term tests, carbon impregnated with potassium iodide and iodine performed better than unimpregnated carbon, and its use dominated early iodine control technology. A water extract from finished impregnated carbons varied in pH from neutral to acidic depending on the method of preparation. As the pH of the water extract of the base carbon also influences the pH of the impregnated carbon, the choice of vegetable-base (coconut shell) carbons for impregnation was helpful because, in addition to being hard, such carbons contain approximately 1 percent potassium hydroxide or sodium hydroxide that reacts with free elemental iodine to produce iodide forms that migrate through the carbon less easily than elemental iodine.

In the late 1960s and early 1970s, researchers realized that design data derived from short-term experiments with fresh carbons provided inadequate adsorber designs for the long-term protection needed from carbon beds. Carbons deteriorate from long exposure to air pollutants (weathering), as well as inadvertent adsorption of widely used organic-compound-containing materials (poisoning; e.g., paint or solvent vapors). Both situations result in a loss of capacity for iodine species. Such observations led to development of deep-bed adsorbers constructed with 4- to 20-inch-deep beds of impregnated carbon that could be filled by pouring the granules into large panels, thereby eliminating the many leak paths associated with tray-type units. This adsorber design was designated a Type III Adsorber. [Note: As of this writing, an addition to the AG-1 Code¹¹ is being developed that will address Type IV Adsorbers, which are similar to Type I Adsorbers.]

The nuclear reactor post-accident iodine release concepts that became established and codified during the late 1960s were based on the assumption that a large quantity of elemental iodine would be released and would

have to be adsorbed. The design criteria were based on the release of 50 percent of core iodine with half of the released iodine captured by plate-out on surfaces. Of the remaining airborne iodine, 85 percent would be elemental, 10 percent would be organic, and 5 percent would be particulate. Contemporary transport concepts contemplated a need to treat large air volumes at locations several steps away from the point of release of the iodine fission products. It was anticipated that iodine capture would be made more difficult by dilution in a large volume of air, as well as by the presence of a large quantity of other chemicals in the air that would compete with iodine for adsorption sites or react more rapidly with the impregnants.

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