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Aging Assessment of Nuclear Air-Treatment System HEPA Filters and Adsorbers

Phase I

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Operated by
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Prepared for
U.S. Nuclear Regulatory Commission

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Abstract

A Phase I aging assessment of high-efficiency particulate air (HEPA) filters and activated carbon gas adsorption units (adsorbers) was performed by the Pacific Northwest Laboratory (PNL) as part of the U.S. Nuclear Regulatory Commission's (NRC) Nuclear Plant Aging Research (NPAR) Program. Information concerning design features; failure experience; aging mechanisms, effects, and stressors; and surveillance and monitoring methods for these key air-treatment system components was compiled. Over 1100 failures, or 12 percent of the filter installations, were reported as part of a Department of Energy (DOE) survey. Investigators from other national laboratories have suggested that aging effects could have contributed to over 80 percent of these failures. Tensile strength tests on aged filter media specimens indicated a decrease in strength. Filter aging mechanisms range from those associated with particle loading to reactions that alter properties of sealants and gaskets. Low radioiodine decontamination factors associated with the Three Mile Island (TMI) accident were attributed to the premature aging of the carbon in the adsorbers. Mechanisms that can lead to impaired adsorber performance include oxidation as well as the loss of potentially available active sites as a result of the adsorption of pollutants. Stressors include heat, moisture, radiation, and airborne particles and contaminants.

Contents

Summary

Nuclear air-treatment systems are one of a variety of systems that provide a safe environment for plant personnel and equipment during normal operations as well as retain radionuclides during accidents. High-efficiency particulate air (HEPA) filters and activated carbon gas adsorption units (adsorbers), designed to capture radioactive particulate and gaseous contaminants, are the key components of these systems. This report presents the results of a Phase I aging assessment for these two key components. Information concerning design features and construction; failure experience; aging mechanisms, effects, and related stressors (the agents or stimuli that can result in degradation); and inspection, surveillance, and monitoring methods (ISMM) are included in the report.

Information is summarized from the standards, codes, and specifications that combine to provide the basic physical, chemical, test, and performance criteria for the glass fiber filter media (paper) and impregnated activated carbon as well as for assembled components. Failure experience, based on analyses of over 60,000 licensee event reports (LERs) and a survey of Department of Energy (DOE) sites, is summarized and, where possible, related to aging (Moeller 1979, Moeller and Sun 1983, Jacox 1989, Sommer and Otermat 1992, and Carbaugh 1983). Although less than one percent of the LERs appeared to be related to filters and adsorbers, several instances of the premature aging of carbon, from error-induced conditions, were reported. Over 1100 failures, or a total of 12 percent of the filter applications, were reported as part of the DOE survey (Carbaugh 1983). Investigators from other national laboratories have suggested that the effects of aging could have contributed to over 80 percent of these filter failures (Johnson et al. 1989). These same investigators reported results of an experimental evaluation of the tensile breaking strength of aged filter media specimens that revealed that 42 percent of the samples did not meet the specifications for test specimens obtained from new material. A significant loss in water repellency from values specified for new media was also measured.

Low radioiodine decontamination factors associated with the Three Mile Island (TMI) accident were attributed to the premature aging of the carbon in the adsorbers. Inspection, surveillance, and monitoring methods have been established to observe filter pressure drop buildup, check both HEPA filters and adsorbers for bypass, and determine the retention effectiveness of aged carbon. Exemptions to TMI technical specifications postponed the latter, surveillance tests that could have revealed the extent of carbon aging.

Aging mechanisms associated with filters range from particle loading of the media to corrosion of metal members, and physicochemical reactions that alter properties of sealants, gaskets, and water repellents. Filter media may embrittle from prolonged exposure to air. In the case of adsorbers, it has long been established that aging mechanisms lead to impaired performance in terms of the capture of volatile iodine species. The deterioration in performance is caused by oxidation as well as the competitive loading of other airborne constituents. Many airborne constituents, including moisture, can readily react or be adsorbed by carbon beds reducing the number of active "sites" that otherwise would be available for radioiodine adsorption. Stressors include heat, humidity, radiation, and airborne contaminants and pollution.

The Phase I study found that the HEPA filters and adsorbers are considered to have a long service life, especially the filters. Thus, if a severe accident happens it is likely to occur at a time when these two final confinement barriers have been in use for an extended period, even years. Even with existing ISMM, the aged, and possibly degraded components, could fail to provide the controlled environment needed by personnel to ensure safe shutdown following the accident, or be the weak link that allows the release of radionuclides to the environment. Further, the Phase I assessment has revealed the need for an improved definition of accident conditions and the possible need for additional information to comprehensively evaluate the performance of aged components under such conditions. The expansion of the Phase I aging assessment to other components of nuclear air treatment systems, namely demisters,

heaters, coarse filters, fans or blowers, and dampers, is recommended. Several of these components, although not designed to retain radionuclides, could mitigate the impact of conditions that threaten to cause failure of aged HEPA filters and adsorbers during accidents.

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Nomenclature

ACS	air cleaning system
ANSI	American National Standards Institute
ASME	American Society of Mechanical Engineers
ASTM	American Society for Testing and Materials
CFR	Code of Federal Regulations
CsI	cesium iodide
DBA	design-basis accident
DF	decontamination factor
DOD	Department of Defense
DOE	Department of Energy
DOP	dioctyl phthalate
EPRI	Electric Power Research Institute
ESF	engineered safety feature
GDC	general design criteria
HEPA	high-efficiency particulate air
ISMM	inspection, surveillance, and monitoring methods
KI	potassium iodide
LER	licensee event report
LOCA	loss-of-coolant accident
MEK	methyl ethyl ketone
MIL	military standard
NEA	Nuclear Energy Agency
NPAR	Nuclear Plant Aging Research

NRC	Nuclear Regulatory Commission, U.S.
OECD	Organization for Economic Cooperation and Development
PNL	Pacific Northwest Laboratory
RG	regulatory guide
RH	relative humidity
SGTS	standby gas treatment system
SSC	systems, structures, and components
STCP	Source Term Code Package
TEDA	triethylenediamine
TMI	Three Mile Island
USAEC	U.S. Atomic Energy Commission

1 Introduction

This report presents the Phase I aging assessment of the high-efficiency particulate air (HEPA) filters and gas adsorption units (adsorbers) of nuclear air-treatment or cleaning and ventilation systems. The study was performed by Pacific Northwest Laboratory (PNL)[a] for the U.S. Nuclear Regulatory Commission (NRC). Information concerning the design and construction of the two air-treatment system components is provided including summaries of related sections of the NRC regulatory guides (RG), American Society of Mechanical Engineers (ASME) standards and codes, and military and American Society for Testing and Materials (ASTM) specifications. These documents combine to provide basic physical, chemical, test, and performance standards for filter media and impregnated activated carbon as well as for assembled components.

Aging mechanisms and effects are discussed in conjunction with stressors, the agents, or stimuli that can result in degradation. Failure experience, based on analyses of licensee event reports (LERs) and a survey of United States Department of Energy (DOE) sites, is summarized and, where possible, related to aging. The discussion of failure experience also includes exhaust-air treatment experience associated with the Three Mile Island (TMI) accident. Although TMI treatment systems provided a significant final barrier to radioactivity release, lower than expected radioiodine removal was attributed to "...poor performance from aged charcoal filters" (Wilhelm and Deuber 1991).

Inspection, surveillance, and monitoring methods (ISMM) are reviewed. These methods are used to establish the condition of the filters and adsorbers once the components have been placed in operation. Surveillance tests, which are a series of tests periodically performed to monitor component condition and demonstrate the current ability to remove fine particles and iodine and iodine compounds, are described along with summaries of related sections of NRC regulatory guides and ASME standards.

Definitions related to aging that are used in the report were obtained or derived from the results of a study to provide uniform common terminology for addressing aging of systems, structures, and components (SSC) in nuclear power plants (EPRI 1992). Aging from normal conditions assumes properly-designed, fabricated, installed, operated, and maintained SSC and is defined as the "general process in which characteristics of [the] SSC gradually change with time or use." It should be noted, however, because of the variety of normal conditions and stressors encountered, when considering HEPA filters and adsorbers, the use of gradually to describe change may not always appropriately reflect the rate at which aging can occur. Normal aging degradation is often the result of exposure to airborne contaminants and concentrations and properties fluctuate. Further, contaminants and pollutants vary from site-to-site and can result from maintenance and testing and possibly even standby or shutdown as well as normal operation. Filters and adsorbers are also used in several systems and large air flows can be involved. Therefore, general rules concerning design service life associated with even normal conditions are essentially impossible to develop. A service life of several years is not uncommon, but significantly shorter periods may be involved, despite the fact that protective components, namely prefilters, demisters, and air heaters, are used to extend usable life. Error-induced conditions leading to early aging are also discussed in the report. Although wearout, including reduced retention efficiency, from error-induced aging degradation can be viewed as premature aging, the root cause of failure in this case is human error, not aging.

[a]Operated for the U.S. Department of Energy by Battelle Memorial Institute under Contract DE-AC06-76RLO 1830.

2 Background

This Phase I aging assessment of HEPA filters and gas adsorption units (adsorbers) is part of an evaluation of the effects of aging on engineered safety feature (ESF) systems, one of the groups of systems of current interest in the Nuclear Plant Aging Research (NPAR) Program (USNRC 1991). The identification of potential safety-related aging issues, coupled with the need to avoid duplication, were the key considerations in identifying candidate ESF systems for initial study. Air-treatment or cleaning and ventilation systems were ultimately selected because failure of these systems can impact both plant and public safety. System components can be the last barrier in preventing the release of radioactivity to the public following an accident including that associated with the airborne iodine and cesium radionuclides that can provide a substantial contribution to total dose. Further, air-treatment systems are needed to ensure safe shutdown of the plant or to allow equipment to be serviced. Satisfactory performance of certain air-treatment systems is essential to ensure control room habitability. Air-treatment systems and components are also used to provide a safe and/or controlled environment for personnel and equipment during normal operations. Operability of some safety-related equipment is dependent upon particular systems to remove heat from the rooms where these components are installed.

As shown in Figure 2.1, air-treatment systems consist of some or all of the following components: moisture separators or demisters, electrical heaters, prefilters, HEPA filters, adsorbers, postfilters, fans or blowers, ductwork, and dampers and valves. Not shown are the instrumentation and equipment to sample and monitor system performance. The first step in the Phase I study was to identify a boundary to isolate the components that would be assessed. The fundamental components in terms of providing plant and public safety are those designed to capture radioactive gaseous and particulate contaminants, namely the adsorbers and HEPA filters, respectively. One or both are installed in nuclear air-treatment systems. The particles could be radioactive chemical compounds or otherwise inert airborne material contaminated by radioactive species. Gasses of primary interest include the elemental and organic forms of radioiodine. Activated carbon is used to remove the gaseous or volatile forms of iodine and is usually impregnated with other chemicals to enhance the removal of the organic species. HEPA filters and activated carbon adsorption units were selected as the focus for initial Phase I assessment not only because they are essential for safety, but in the case of the adsorbers, it has long been established, and was evidenced again during the TMI accident, that there are aging mechanisms that can lead to impaired performance (Burchsted et al. 1976, Wilhelm and Deuber 1991).

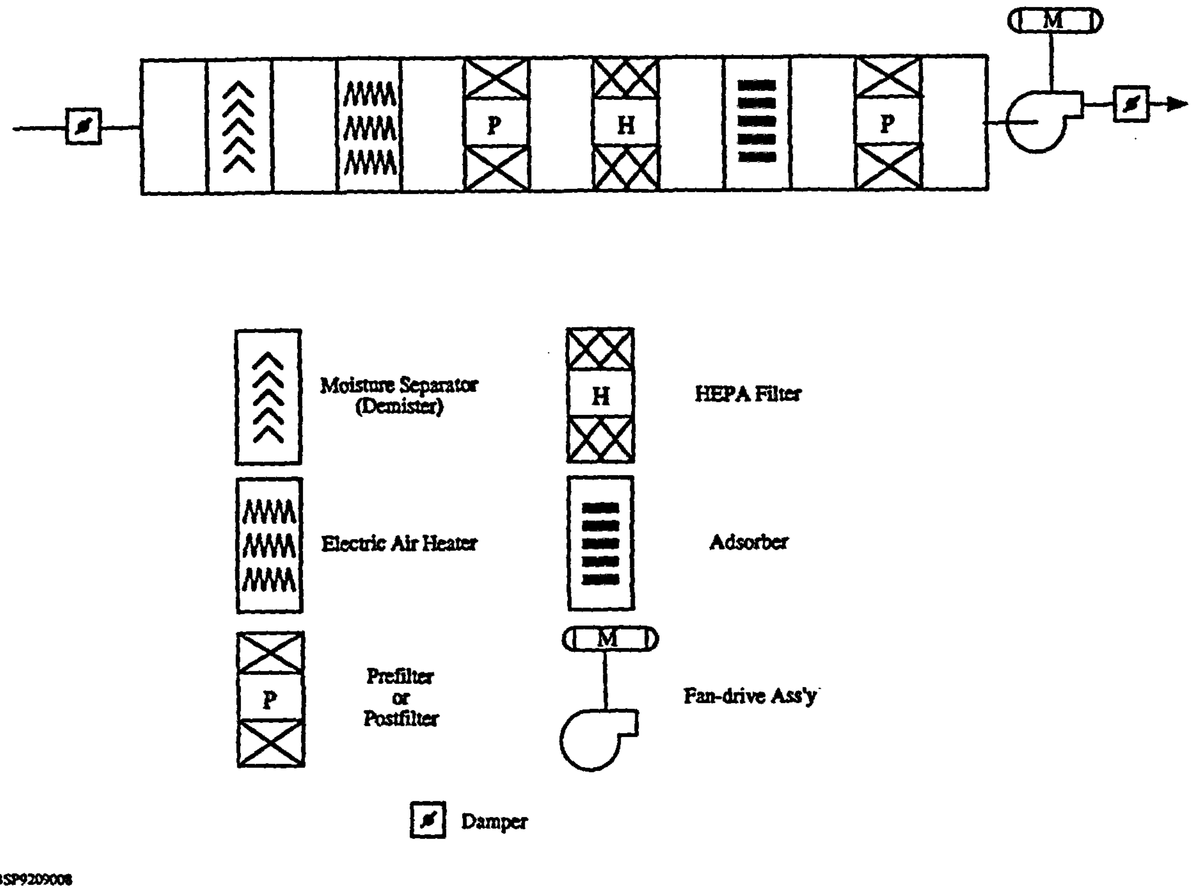

Figure 2.1. Common Air-Treatment System Configuration

3 Component Design and Construction

Particle filtration and gas (vapor) adsorption units (adsorbers) are included in nuclear air-treatment systems and are designed to remove radioactive materials. The materials, airborne particles or volatile species, may be suspended in or exist as gas phase constituents of recirculating aerosols, gaseous effluents, or accidental releases. The adsorbers, activated carbon beds, can effectively remove elemental radioiodine and are often impregnated with other chemicals to enhance the retention of organic species. The fine particle filters, HEPA filters, may also be significantly involved in radioiodine removal because iodine can be in the form of solid cesium iodide (CsI). Design and construction of the two components are discussed below.

3.1 HEPA Filters

HEPA filters clean or treat aerosols by separating suspended particles from an essentially atmospheric pressure gas stream. The filter media is made from a mixture of glass fibers and is in the form of filter "paper." Particles are collected by interception, impaction, and Brownian diffusion. The continuous gas phase passes through virtually unchanged. HEPA filters are defined as a "high-efficiency particulate air filter having a fibrous medium with a particle removal efficiency of at least 99.97% for 0.3 μm particles of dioctyl phthalate [DOP]" (ASME 1989a).

An excellent review of the construction and service characteristics of HEPA filters has been prepared by First (1991). This reference also includes a brief history of the development of filters for the nuclear industry and includes additional details concerning filtration mechanisms. Much of the information presented in this section was either abstracted or taken directly from First or referenced military specifications. As indicated by First, "Filters constructed with paper pleated the full depth of the rigid outer frame and with adjacent pleats held apart by full-depth corrugated separators are the most widely used." In other words, as shown in Figure 3.1, the glass fiber filter paper is in the form of a continuous sheet pleated vertically over the separators.

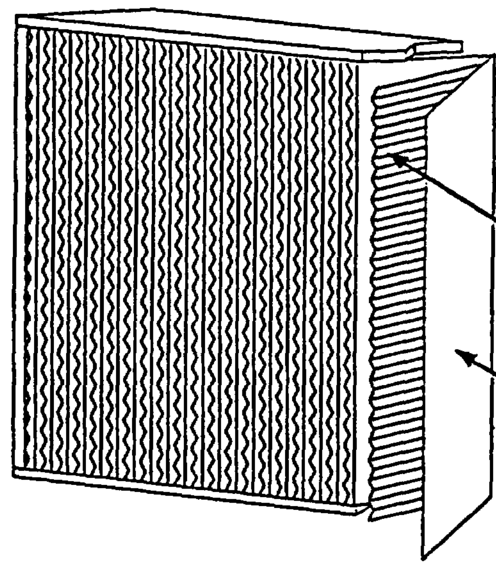

Figure 3.1. HEPA Filter

Physical and chemical filter medium/paper standards are contained in Military Specification MIL-F-51079D, "Filter Medium, Fire-Resistant, High-Efficiency," and include those for air flow resistance, particle penetration, and tensile strength (U.S. Department of Defense [DOD] 1985). The latter, tensile strength criteria, includes requirements after exposure of the filter paper to heated air, water immersion, and after exposure to gamma irradiation. Standards for water repellency prior to and after exposure to gamma irradiation and mildew resistance are also included. The penetration criteria (not to exceed 0.03% for 0.3 μm DOP particles) is directly related to the minimum efficiency used in the above definition. The penetration criteria is associated with tests specifying a flow rate of 0.032 m^3/min and an exposed test area of 100 cm^2. Maximum pressure drop related to the preceding test specifications (minimum velocity of 320 cm/min) is 0.39 kPa (40 mm of water). The minimum average tensile breaking strength specified for media specimens (N/m or lb/in. of width in the cross direction), initially, after exposure to heated air at 371°C ± 28°C (700°F ± 50°F) for 5 min, after being soaked for 15 min, and after radiation exposure are 350 or 2.0 (438 or 2.5 in the machine direction), 105 or 0.6, 175 or 1.0 (wet) and 175 or 1.0, respectively. The military specification also includes the requirement that the combustible material in the filter medium shall not exceed 7 wt%. First (1991) notes that the latter "permits incorporation of... organic matter, divided between (1) a latex addition to give the paper strength and resistance to cracking at the bends [where the sheet is pleated] and (2) a water repellent to protect the paper against wetting from deposition of liquid droplets, should they be present." Specifications for the filter medium, derived from MIL-F-51079, are also presented in the ASME Code on Nuclear Air and Gas Treatment, ASME/American National Standards Institute (ANSI) AG-1-1988, Section FC, "HEPA Filters" (ASME/ANSI 1988).

In addition to the medium, HEPA filter components include the frames, gaskets, face guards, and the corrugated separators that are used to separate adjacent sheets of filter paper. Specifications for these components are presented in MIL-F-51068F, "Filters, Particulate (High-Efficiency Fire Resistant)," along with criteria for the adhesives, sealants, and paints used to assemble and finish the units (U.S. Department of Defense 1986). A rigid outer frame or housing is used to fully enclose the paper-separator assembly on four sides, and provide the connections to adjacent ductwork. Specifications for frame material include those for plywood, wood particle board, laminated veneer lumber, aluminum alloys, cold-rolled steel sheet, chrome steel sheet, and stainless steel. A flame spread classification is given for plywood. First (1991) notes that "face gaskets are usually constructed from flat strips of expanded closed cell neoprene sponge rubber... cork and other rubber formulations are also permitted [a proprietary fluid seal design is also discussed]." It is specified that the corrugated separators be made from aluminum alloys or epoxy-coated aluminum. The specification requires that "adhesives used to splice the medium, fasten the gasket to filter face, and sealants or adhesives used to seal the filter pack to the frame shall be self-extinguishing after direct contact with an open flame..."

Performance specifications for assembled standard-sized filters (nominal rated capacity ranging from 0.71 to 57 m^3/min or 25 to 2000 ft^3/min) are included in MIL-F-51068F for DOP particle penetration (no greater than 0.03% at specified flow rates) and resistance to air flow, pressure, heated air, and environmental exposure. Maximum pressure drop specified at rated air flow ranges from 0.25 to 0.32 kPa (1.0 to 1.3 in. of water), depending on size. The pressure test for 61 by 61 cm units involves assuring that the assembled unit, after conditioning for not less than 24 hours in a chamber at 35°C (-95°F) and at a relative humidity (RH) of 95%, will then withstand a specified air flow (needed to produce a 2.49 ± 0.05 kPa or 10 ± 0.2 in. water pressure differential across filter) and water spray environment without rupture of the filter medium. Evaluation of environmental performance involves cyclic exposure for a total of 9 weeks to specified conditions termed arctic (-54°C or -65°F), desert (45°C or 160°F and 10% RH) and tropical (45°C or 113°F and 88% RH). Particle penetration criteria also include minimum values for the wet filter following the pressure test, after exposure to specified flow rates of heated air (371 ± 28°C or 700 ± 50°F for at least 5 min), and after the environmental exposure cycles.

Materials, design, and fabrication standards are also included in Section FC of the ASME Code on Nuclear Air and Gas Treatment (ASME/ANSI 1988). Three types of HEPA filters are addressed: folded filter media with corrugated separator/supports (described above); minipleat medium with glass fiber or noncombustible thread separators (made from a series of flat panels of pleated filter media assembled in a V-shape, with adjacent pleats separated and supported by ribbons of glass fiber or noncombustible threads glued to the media, maximum pleat height of 20 mm); and continuous corrugated filter media folded without separators. Steel and stainless steel sheet frames are emphasized. A sequence of qualification tests is described that includes evaluation of resistance to air flow, particle penetration, resistance to rough handling, resistance to pressure, resistance to heated air, and spot flame and radiation resistance and involves a specified number of filters. No attempt was made to define all differences between the above military specifications and Section FC, but it was noted that combustible material in the filter media is limited to 5 wt% by the latter.

3.2 Adsorbers

As indicated above, adsorbers are used to remove gaseous radioiodine species from effluents. Various standards and specifications permit the use of any adsorbent medium that has been demonstrated to be equal to or better than activated carbon in terms of radioiodine retention. However, only activated carbon is discussed because it is essentially the only material used and supporting testing procedures and acceptance criteria are specific to its use.

Gasses are directed through tightly-packed beds of activated carbon granules. The carbon or charcoal is prepared by controlled heating in a steam environment to remove volatile organic material. The heating generates a material with a large surface-to-mass ratio and internal surfaces or sites where adsorption of iodine molecules can take place. The active media is usually "... activated charcoal that is derived from either coal or coconut shells. Although elemental radioiodine is retained efficiently by activated carbon alone ..., the charcoal is often impregnated with additional chemicals to improve its retention of organic species (e.g., methyl iodide). Typical impregnants include potassium iodide (KI), triethylenediamine (TEDA), or derivatives of TEDA. Mixtures of these compounds are also used.... With KI as the impregnant, iodine retention is mainly due to isotopic exchange [nonradioactive iodine in the impregnant substitutes for the radioactive iodine of the organic species and then passes through the bed]... With TEDA or its derivatives as the impregnant, the primary retention reaction is due to the formation of quaternary salts. Even in this case, however, isotopic exchange is an important factor..." (Wilhelm and Deuber 1991).

Standards for the adsorbent media are included in the "Code on Nuclear Air and Gas Treatment," ASME/ANSI AG-1-1988, Section FF, Adsorbent Media (ASME/ANSI 1988). In addition to specifications for physical properties, Section FF lists requirements for testing the adsorbent to verify retention performance. A free-flowing granular substance is specified. Activation and impregnation processes are left to the discretion of the manufacturer. Physical tests specified after impregnation include those for apparent density, particle size distribution, ignition temperature, and hardness. Qualification tests to establish the suitability of a grade or type of impregnated activated carbon require verification that elemental iodine removal efficiency is not less than 99.9% at 30°C and 95% RH. Corresponding efficiency values for methyl iodide are 99.0% at 80°C and 95% RH and 98% at 130°C and 95% RH. Further, batch tests at the time of manufacture, to assure that the material being used has the same characteristics as the material qualified in testing, require demonstration of minimum elemental iodine and methyl iodine retention efficiencies of 99.5% (at 180°C) and 97% (at 30°C and 95% RH), respectively. Definitions and terms included in Section FF include those for "batch," "grade or type," and "qualification test."

ASTM standards are referenced in Section FF of AG-1-1988 that describe methods for testing the adsorbent with radioactive material to verify retention characteristics as well as to evaluate physical properties such as density and particle size distribution. The former, ASTM D 3803-89, "Standard Test Method for Nuclear-Grade Activated Carbon," is "... a very stringent procedure for establishing the capability of ... activated carbon to remove radio-labeled methyl iodide from air and gas streams" (ASTM 1989). As will be discussed later in the report, the test method is used to quantify the extent of degradation of aged carbon as well as to qualify new carbons. Physical properties and performance specifications for new, virgin impregnated activated carbon for removing gaseous radioiodine are specified in ASTM D 4069-90, "Standard Specification for Impregnated Activated Carbon Used to Remove Gaseous Radio-Iodines from Gas Streams" (ASTM 1990). Test methods for evaluating the various physical properties are included as referenced documents.

Specifications for materials, design, fabrication, and the inspection and testing of two types of adsorbers are presented in Sections FD, "Type II Adsorber Cells," and FE, "Type III Adsorbers," of ASME/ANSI AG-1-1988 (ASME/ANSI 1988). Type 304 or 304L series stainless steels are specified for materials that contact the adsorbent. General design of the Type II adsorber unit is based on a single cell consisting of 50.8-mm (2-in.) thick minimum beds in a modular tray-type arrangement. A minimum residence time of 0.25 sec. and maximum pressure drop of 0.31 kPa (1.25 in. water) at the rated capacity of 9.43 m^3/min (333 ft^3/min) is specified. Type III design is based on single or multiple permanent units where carbon is filled and emptied in place (sometimes called deep bed or gasketless, see ASME/ANSI AG-1-1988 Section FE for additional details).

3.3 Supporting Documents

In summary, basic qualifications for the filter medium and assembled HEPA particulate filters are contained in two military specifications (U.S. Department of Defense 1985, 1986). Primary standards for adsorbent media, assembled adsorbers, and related testing methods are presented in Sections FD, FE, and FF of ASME/ANSI code AG-1 and two ASTM specifications (ASME/ANSI 1988; ASTM 1989, 1990). Numerous documents supplement and/or support the preceding five references, including NRC and other ASME and ASTM publications. One of the most closely related NRC publications, RG 1.52, "Design, Testing, and Maintenance Criteria for Post Accident Engineered-Safety-Feature Atmosphere Cleanup System Air Filtration and Adsorption Units of Light-Water-Cooled Nuclear Power Plants," is currently being revised (USNRC 1978; Bellamy 1991). This guide presents methods for implementing the appropriate general design criteria (GDC) described in Appendix A of 10 CFR 50 (U.S. Code of Federal Regulations 1992) and as suggested by the title of the guide, "... applies only to post accident engineered-safety-feature atmosphere cleanup systems designed to mitigate the consequences of postulated accidents ... [and] does not apply to atmosphere cleanup systems designed to collect airborne radioactive materials during normal plant operation, including anticipated operational occurrences." Regulatory Guide 1.140, "Design, Testing, and Maintenance Criteria for Normal Ventilation Exhaust System Air Filtration and Adsorption Units of Light-Water-Cooled Nuclear Power Plants" provides guidance for the latter, normal systems (USNRC 1979).

Regulatory Guide 1.52 references two ANSI/ASME standards for the design and testing of engineered safety feature (ESF) system components including HEPA filters and adsorbers. The latest revisions of these two standards have been published by the ASME as American National Standards ASME N509-1989, "Nuclear Power Plant Air-Cleaning Units and Components," and ASME N510-1989, "Testing of Nuclear Air Treatment Systems" (ASME 1989 a,b). These two ASME standards, in addition to being connected to the regulatory guides, are also at least the initial link to the basic five references that contain various construction, material, test, and qualification specifications for new HEPA filters and adsorbers. Specifically, requirements of ASME N509-1989 include the use of the two military specifications and the three sections of the nuclear air and gas treatment code (the above two ASTM standards are referenced in Section FF of code, ASME/ANSI AG-1). There is no intent to imply that all of the information presented or referenced in the latest revisions of the two ASME standards has been evaluated by the NRC in terms of applicability or acceptability. Further, the use of the term requirement in conjunction with the various standards, codes, and specifications does not necessarily mean that a particular requirement has been accepted by the NRC. Finally, it should be noted that some of the information included in the two ASME standards and NRC regulatory guides will be discussed in greater detail later in the report in conjunction with surveillance tests that are used to monitor the condition of installed filters and adsorbers.

4 Aging Mechanisms, Effects, and Stressors

This section combines information concerning aging mechanisms and effects and the various stressors that are involved. As will be detailed below, stressors, the agents or stimuli that can contribute to aging (including the direct degradation in performance in terms of radionuclide retention) include heat, humidity, steam, radiation, and airborne contaminants and pollutants.

4.1 HEPA Filters

The obvious effect of service associated with HEPA filter use is the increased pressure drop (resistance to air flow) through the filter media arising from particle retention (loading). As indicated in Section 3.0, new or clean filters are designed to provide a resistance of about 0.25 kPa. First (1991) notes that "a dust pickup of approximately 1 kg/1000m^3/h of design filtration capacity represents a resistance increase of 0.25 kPa from a new condition." In a relatively clean environment, filters may be used for several years before replacement is required. Dust loading, along with heat and radiation, also have the potential to reduce the effectiveness of the organic materials used to strengthen the filter medium and provide water repellency.

High moisture content is another stressor that can lead to increased pressure drop as well as reduced filter medium strength. Ricketts, Ruedinger, and Wilhelm (1987) note that "Moisture induced . . . [deterioration] in filter performance and filter structure failure result principally from the presence of liquid water in the fiber structure of the filter medium." These authors discuss incorporation of liquid water into the filter medium by sorption, condensation, or droplet filtration (interception) and also summarize a large number of other literature sources that deal with moisture effects on filter media and construction materials and assembled units. In conclusion, they note that ". . . water in the filter medium leads to an increase in differential pressure and to . . . [deterioration] in filter pack stability and in filter medium performance characteristics, especially the tensile strength. The mechanical load on the filter is thus increased at the same time that the structural strength is decreased. The end result is that filter structural failure can occur for unacceptably low values of ΔP, even at design flow rates. . . ." (Ricketts, Ruedinger, and Wilhelm 1987). In reporting the results of an experimental program these same authors note that "the 21 filters tested at 50°C, constant volumetric air flow rate, and ambient pressure demonstrated that the differential pressure of new clean filters increased significantly only above 95% RH, rose up to 0.5 to 2 kPa at 100% RH and reached values between 6 and 9 kPa during filtration of liquid-water aerosols. . . ." (Ruedinger, Ricketts, and Wilhelm 1985). Additional information concerning the effects of moisture, emphasizing immediate failure rather than gradual deterioration and including details related to dust-loaded filters, is presented later in this report.

Filter media may embrittle from prolonged exposure to air containing normal concentrations of oxygen and oxides of nitrogen and sulfur. Aging mechanisms or processes that can gradually change the physical characteristics of HEPA filter components other than the media include corrosion of metal members and physicochemical reactions that alter the properties of sealants, gaskets, and water repellents. Metal components that could be affected include the frames and corrugated separators. Stressors associated with physicochemical reactions potentially affecting face gaskets and the adhesives and sealants that are used to splice the medium, fasten the gaskets to the filter face, and seal the filter pack to the frame, include heat and radiation.

Johnson et al. (1989) have reported results of tests to quantitatively evaluate the effect of aging on HEPA filters. In one group of tests aged, flat, sheet media samples (obtained by dismantling filters with 13 to 14 years of service) were tested using the equipment and procedures of MIL-F-51079 (Department of Defense 1985), the specification discussed earlier that includes the physical and chemical criteria for the media used in new filters. In summary, 42% of the samples failed tensile strength tests; i.e., the samples did not meet specifications for new media. Johnson et al. also note that "due to the brittleness of the bend area of the pleat, a sample could not be obtained for testing. There is no doubt, however, that this area of the aged HEPA filter media represents the weakest part and it should have even lower tensile strength value." The bench-scale tests also revealed a significant loss of water repellency. All samples failed when the top or dirty side of sheets were tested while four of the seven samples failed to meet the requirements for water repellency when the bottom or clean side was examined. As anticipated, due to service, 71% of the samples failed the pressure drop test. However, all samples passed the DOP efficiency tests (met the 99.7% requirement for new material).

In addition to the above bench-scale tests, several of the aged filters were exposed in a wind tunnel to generate pulses equal to those associated with the standard NRC Region I design basis tornado, and using a shock tube, to shock wave overpressures. Six filters, in service for 15 to 19 years, and two filters, in service for 14 years, were subjected to the tornado pulse and additive shock overpressure tests, respectively. Johnson et al. report that "the average breaking pressure of the six aged filters was 1.38 ± 0.95 psi [9.51 ± 6.6 kPa]. The comparable breaking pressure of a new unused filter is 2.89 psi [19.9 kPa] The average breaking pressure of the two aged filters utilizing small incremental pressure increases is 1.8 psi [12.4 kPa]. The comparable breaking pressure of a new unused HEPA filter is 2.5 psi [17.2 kPa]. . . ." In addition to these 52% and 28% decreases in breaking pressure, a large increase in complete filter pack blow-out was reported for the aged filters during the simulated tornado tests.

4.2 Adsorbers

Exposure to air containing stressors, namely moisture and contaminants or pollutants, can slowly and continuously degrade the performance of gas adsorbers with time. This aging, also termed weathering, is inherent because of the nature of the material used, i.e., one that has been "activated" to dramatically increase surface to mass ratio and provide countless reaction sites, coupled with the essentially ubiquitous nature of the contaminants. Many airborne constituents, including moisture, can readily react or be adsorbed by carbon beds reducing the number of active "sites" that otherwise would be available for radioiodine adsorption. During normal operation, charcoal may be exposed to air flows containing contaminants including volatile organic solvents, sulfur dioxide, nitrogen oxides, and carbon monoxide. Because of system flow rates, even traces of pollutants can have a significant, cumulative effect. Oxidation, as well as competitive loading, can impair bed performance, including decreasing the efficiency of the impregnant. Because airborne constituents can vary dramatically with time and from site-to-site, it is essentially impossible to provide criteria concerning the useful life of impregnated activated carbon.

Aging degradation or weathering of iodine adsorbers as a result of exposure to various stressors has been treated extensively in the literature, particularly in terms of methyl iodide retention efficiency. Results of numerous experimental investigations involving the latter compound have been reported. Two summary reviews, which also include numerous pertinent references, have been prepared by Wilhelm and Deuber (1991) and a group of Nuclear Energy Agency (NEA) experts [Organization for Economic Cooperation and Development (OECD) 1984]. Briefly, studies have revealed that increasing temperature and humidity can be agents that cause aging and aging effects. Deitz (1978) notes that ". . . there is strong evidence that the interaction of water vapor and charcoal is a significant factor in the degradation of the charcoals when the RH is 70% and greater. The laboratory air mixtures studied [in this investigation of methyl iodide penetration] were water vapor, water vapor and sulfur dioxide, water vapor and ozone, and water vapor and carbon monoxide. . . ."

Wren and Moore (1991a) in an investigation of the adsorption and desorption behavior of contaminants studied the effect of dry and humid conditions on adsorption of NO_2, SO_2, methyl ethyl ketone (MEK), and NH_3. Their conclusions concerning the latter, humid conditions include the observations that ". . . adsorbed water increases the adsorption rate and capacity of TEDA charcoal for NO_2, while it does not significantly change those for SO_2. However, it appears that SO_2 is adsorbed as H_2SO_4 on the wet charcoal. Adsorbed water slightly reduces the adsorption capacity of the charcoal for MEK, but does not affect the adsorption of NH_3." These same authors, reporting on the effect of contaminants on CH_3I removal efficiency, noted "The efficiency of TEDA charcoal is degraded most by NO_2 and SO_2, NH_3 has a negligible effect, and MEK produces a mild degradation. The degree of degradation parallels the contaminant's ability to be chemisorbed.... Nitrogen dioxide adsorbed under dry conditions is more effective in degrading the CH_3I removal efficiency of the charcoal than when adsorbed under humid conditions. On the other hand, a completely opposite result is observed for SO_2. The MEK contaminant behaves similarly to SO_2, but the effect of humidity was less significant than for SO_2. Ammonia has no effect on the efficiency of the charcoal regardless of humidity" (Wren and Moore 1991b).

Amine (TEDA or derivatives to reduce volatility) impregnated carbons age less rapidly than those where KI is used or that are unimpregnated (OECD 1984). In an investigation of temperature and humidity on the aging of TEDA impregnated charcoals, Billinge and Broadbent (1989) note that "... KI impregnated charcoals ... rapidly lose efficiency when exposed to high humidity air [and] this deterioration was mainly caused by oxidative aging of the carbon surface ... Amines are a class of compounds well-known for their anti-oxidant properties so TEDA carbon may age more slowly than KI material because the rate of formation of organo-oxygen surface groups is reduced; a second possibility is that the basic properties of the amine neutralize any surface acidity formed...."

The effect of the accumulation of organic contaminants on the aging process was investigated by Hyder (1989). As in the case of the above investigation involving MEK, results "... show the effect of organics on carbon performance [in terms of methyl iodide] is limited ... Some inference is possible regarding the mechanism by which sorbed organics affect methyl iodide retention. In these used carbon samples, all of the impregnant TEDA has been lost by evaporation or reaction. Methyl iodide retention depends on the availability of the potassium iodide impregnant for exchange with the methyl iodide. Other aging processes chemically alter this impregnant or remove it, but the sorbed organics do neither...."

As indicated above, much of the literature concerning aging mechanisms and effects is related to methyl iodide penetration. It is noted in an older but valuable and comprehensive review of air cleaning systems (ACS) (Burchted, Kahn, and Fuller 1976) that "the loss in capacity for elemental iodine is much slower than that for methyl iodide. Beds exposed continuously to flowing air at one installation showed adequate remaining capacity for elemental iodine after 4 years of service. At other installations, however, exposure of beds to paint and solvent fumes reduced capacity to the point that efficiency fell to unacceptable levels in only a few months...." Adequate long-term performance in terms of removing elemental iodine from plant ventilation exhaust air under normal operating conditions is also reported in Pelletier et al. (1978). The associated filter systems were used continuously for treating effluents from the auxiliary building. Average efficiency over a 3-year period was 99.23%.

Relatively rapid deterioration of stainless steel components can result from contact with wet carbon. Severe pitting resulting from galvanic corrosion has been reported (Liening 1991). Obviously, carbon should be removed immediately if it becomes wet. The revised NRC RG 1.52 "... will clearly indicate that wetting of impregnated activated carbon should be avoided because it establishes conditions for rapid chemical corrosion between the impregnants and stainless steel structural material supporting the charcoal beds" (Bellamy 1991).

Finally, it should be emphasized that the possibility of impaired performance in terms of radioiodine retention as a result of the aging or weathering of impregnated activated carbon has been a concern almost from the start of the nuclear industries' existence. In fact, concern about the useful life of carbon could be considered one of the first, if not the first, aging issue. As will be detailed later in the report, the issue is addressed by a surveillance test, a series of tests that are periodically conducted to define the remaining radioiodine retention capacity of used or exposed carbon. However, as will also be discussed, additional consideration of aging issues is believed to be warranted for both HEPA filters and adsorbers, especially in relation to accident environments.

5 Failure Experience

This section describes failure experience that has been reported for HEPA filters and adsorbers. Where possible, the experience is discussed in terms of aging effects and degradation and in conjunction with specific stressors. Information related to both normal operating and accident conditions is presented. Analyses of LERs submitted by commercial nuclear power plant operators are reported along with results of a survey of DOE sites to obtain information on the number of and reasons for HEPA filter changeouts and failures. Information to evaluate air-cleaning system performance during the TMI accident is also summarized.

5.1 Licensee Event Reports

An unusually large amount of potentially useful failure data were found in the literature. Analyses of LERs covering the periods from January 1, 1975 through June 30, 1978, 1978-1981, 1985-1987, and 1988-1991, to identify those pertaining to air-monitoring, air-treatment, and ventilation systems, have been prepared by Moeller (1979), Moeller and Sun (1983), Jacox (1989), and Sommer and Otermat (1992). In summary, 15% of 43,500 reports were associated with the subject systems while an estimated, roughly 100, or about 2% of the system reports, appeared to be associated with HEPA filters and carbon adsorbers. No relationship to aging was reported.

Moeller and Kotra (1985) and Moeller and Sun (1987) reviewed LERs for the periods 1981 through 1983 and 1984 through 1986, respectively, to isolate those pertaining to control room habitability. Of the approximately 20,800 reports, 4% were related to this focus. During the 1981 through 1983 period, 28 of the LERs were associated with impaired filter or adsorber efficiency. Moeller and Kotra note that "The majority of LERs related to HEPA filters and charcoal adsorbers reflect that the performance of such units degrades with time and that routine tests reveal that their efficiencies do not meet technical specifications. The normal licensee response is to replace the units. More serious occurrences, however, include the fouling of charcoal adsorbers by welding and/or paint fumes and the occasional wetting of filters or adsorbers by drainage water or the inadvertent actuation of their associated fire protection deluge systems. At one plant, paint fumes contaminated the control room emergency charcoal filters so severely that their measured removal efficiencies were significantly degraded. Apparently, the control room emergency ventilation system was in a recirculation mode during painting in an adjacent room. Backflow through component drain lines into filter packages necessitated their replacement at one plant... Unintentional actuation of fire suppression systems was reported at three... plants... [that] rendered the filters and adsorbers inoperable and they had to be replaced."

Moeller and Sun note that "a total of 30 LERs related to charcoal adsorber problems were filed between 1984 and 1986. Many of these events resulted from tests having revealed that the charcoal adsorber bank associated with an emergency ventilation system was not able to remove the required more than 99% of the halogenated hydrocarbon refrigerant test gas as stipulated by the Technical Specifications [test is discussed later in the report in Section 6.2]. In most cases, the reduction in adsorption capability was thought to be due to moisture having gained access to the charcoal. Compounding the problem was the fact that round robin tests had shown that commercial charcoal testing laboratories, both in the U.S. and abroad, lacked the capabilities of determining the adsorption capacity of such charcoals on an accurate and reliable basis."

5.2 DOE Site Survey

Using a survey, Carbaugh (1983) collected data on the numbers of and reasons for HEPA filter changeouts and failures at DOE sites for the years 1977 through 1979. A total of 1105 filter failures, or 12% of the 9154 filter applications, was reported. The ratio of filters changed to those failed, 6.2 to 1, suggested that most filters are changed out prior to failure. Carbaugh notes that "the largest majority (63%) of filter changeouts were attributed to high differential pressure (ΔP) across the filter, indicative of filter plugging.... The vast majority of filters were reported as [having been] exposed to no distinguishing environmental characteristics (i.e., they filtered essentially clean, dry, air environments similar to those that might be found in typical building ventilation systems or in systems with good pre-HEPA treatment features).... The majority of [the 1105] failures occurred for unknown or unreported reasons.... The incidences of frame failures, gasket or seal failures, and filter media ruptures were approximately equal; each constituted between 5% and 6% of all filter failures. When frame failure was identified, essentially all failed frames (58 out of 65) were wood. No observation of steel frame failure was reported.... calculating the ratios of seal-type failures to seal-type applications [3920 gaskets and 151 fluid seals] can lead one to conclude that little difference may exist between gasket and fluid seal failure rates.... All media ruptures occurred in wood-frame filters, and almost all (48) had aluminum separators and polyurethane foam sealants. [Media to frame] Sealant failure was not identified as a significant filter failure mode (3 occurrences reported out of 1105). ... Ratios of filters failed to filter applications in which these failures were experienced are several times higher for HF acid and high moisture environments than for environments having no distinguishing characteristics, or the average of all single environment exposures [exposure of filters to a single significant environmental factor, e.g., solvent, high moisture, high dust]...."

Johnson et al. (1989) explain the results of the above survey in the following manner: "If one examines the categories reported for filter failure, it is evident that the effects of aging could contribute to 81% of these failures, except for the 19% resulting from handling or installation damage." Johnson et al. also provide two other references that include qualitative information that reveal indications of aging: a technique to dismantle and separate HEPA filter components, based on unfolding the pack and rerolling the media, worked well on new filters but became impractical on used filters because of the "weakness of the filter paper"; and as part of a report concerning the in-service aging effects of unused and used cleanroom, chemical, and radioactive contaminated HEPA filters it was noted that "aluminum spacer deterioration was observed in all used filters examined with the most significant levels observed when high humidities and acid gases were present."

5.3 TMI Accident

As suggested earlier in the report, selection of adsorbers for Phase I study was reinforced by literature references revealing that the low radioiodine decontamination factors (DF) associated with the TMI accident were attributed to the premature aging of charcoal. Before presenting details related to aging issues, it should be emphasized that building ventilation systems played an important role in decreasing the radionuclide release associated with this incident and in fact, prevented the release of most of the radioiodine. The estimated quantity of I-131 released was 5.6×10^{-9} Bq or 15 Ci (in contrast to about 9.3×10^{-4} Bq or 2.5 million Ci of noble gas) (Rogovin and Frampton 1980).

Despite the above, the terms "somewhat dismal results" and "rather unsatisfactory" have been used to describe exhaust air filtering experience during the TMI accident (OECD 1984; Wilhelm and Deuber 1991). In summary, it was determined that the filtering systems installed at the time of the accident provided a DF of 9.5 for all species of radioiodine (corresponds to a retention efficiency of 89.5%), and the radioiodine releases were higher by about a factor of five than they would have been if there had been no system deterioration. The preceding values and reasons for the degradation are detailed in Rogovin and Frampton (1980) and discussed by Bellamy (1981) in a paper that was prepared to summarize pertinent (related to air-cleaning technology) efforts and recommendations of various investigative groups. Briefly, system degradation and the fact that radioiodine retention was associated with an efficiency of 89.5% was ultimately attributed to pre-accident aging of the impregnated activated carbon. For a number of reasons ventilation air was passed through exhaust systems from the time of carbon installation to just shortly after the accident. This approximately 1-year time frame included periods of exposure of the carbon beds to fumes from painting and cleanup efforts. Furthermore, exemptions to technical specifications postponed surveillance tests that could have revealed the extent of aging. (Tests are described in the next section and involve the periodic laboratory evaluation of exposed carbon samples.) The exemptions also permitted the use of carbon with a methyl iodide removal efficiency of 96.97%.

Carbon removed from four filter trains following the accident, along with data on various iodine species obtained from an air sample, suggest removal efficiencies ranging from 49.1% to 75.6% and 98.5% to 99.9% for methyl iodide and elemental iodine, respectively. These efficiencies are based on calculated releases conservatively assuming 95% RH and percentages of volatile species of 40% and 35% for organic and elemental iodine, respectively. The efficiency range is attributed to unbalanced ventilation flows. It should be noted that it was estimated that the highest RH that existed inside the auxiliary building during the accident was 80%.

Review indicated that releases of radioactive material in particulate form were negligible. Postaccident efforts included changeout of HEPA filters as well as carbon. As indicated in Rogovin and Frampton (1980) "These HEPAs were visually examined before changeout and were intact and in satisfactory condition, but were damaged during changeout.... Unfortunately, no used HEPA filters or sections of filter media were retained for analysis." Again, the highest RH estimated for the auxiliary building was only 80%.

6 Inspection, Surveillance, and Monitoring Methods

This section describes the inspection, surveillance, and monitoring methods (ISMM) that are used to establish the condition of HEPA filters and adsorbers once the components have been placed in operation. As will be detailed below, the use of surveillance tests, series of tests periodically performed to monitor component condition and demonstrate the current ability to remove fine particles and iodine and iodine compounds, is emphasized. Surveillance tests include in-place leak tests and visual inspections to evaluate filter banks and adsorbers in terms of component damage and bypass. Also included, and specifically related to aging concerns, are laboratory tests of used or aged carbon samples to determine remaining radioiodine adsorption capacity. In addition to the surveillance tests, HEPA filter pressure drop is continuously monitored (alarmed and indicated).

6.1 HEPA Filters

Instrumentation provisions, including alarms, for air-cleaning systems and components are presented in ASME N509-1989 (ASME 1989a). High differential pressure alarms are specified for local as well as remote-manned control panel locations for the HEPA filters of ESF air-cleaning systems. Pressure drop indication is specified for the local location. For non-ESF units, differential pressure indication and high differential pressure alarms are recommended only for the local location. Particle loading along with the moisture content of the air most adversely influence the rate and extent of filter pressure drop increase. As indicated earlier, most filters are changed out prior to failure, primarily as a the result of an indication of a high differential pressure across the filter.

Criteria for surveillance tests involving installed systems and the various components are presented in ASME N510-1989 (ASME 1989b). As in the case of new components, other documents, including NRC RG 1.52 and RG 1.140, supplement and support this basic reference (USNRC 1978, 1979). Visual inspection is recommended before each test series and specific guidance is provided for HEPA filters and adsorbers. Air flow distribution tests for both HEPA filter and adsorber banks are described. Testing to verify uniform mixing in the air stream approaching the HEPA filter bank or adsorber stage is a prerequisite for conducting surveillance leak testing of the installed filter bank. The recommended frequency for this "air-aerosol mixing uniformity test" includes performance after completion of initial construction and after major system modification or repair (acceptance tests). The method is based on the introduction of DOP aerosol and "... concentration readings ... taken across a plane parallel to, and a short distance upstream of, the HEPA filter bank or adsorber stage" (ASME 1989b).

Surveillance leak testing of installed HEPA filters is specified because gradual deterioration and leaks could develop under standby as well as service conditions. This test is also based on a DOP challenge aerosol introduced upstream of the filters. Concentrations upstream and downstream are then measured. Recommended frequency given in Table 1 of ASME N510-1989 includes acceptance testing, after each HEPA filter replacement and at least once each operating cycle (ASME 1989b). Nuclear Regulatory Commission RG 1.52 states that "HEPA filters should be tested in place (1) initially, (2) at least once per 18 months thereafter, and (3) following painting, fire, or chemical release in any ventilation zone communicating with the system to confirm a penetration [ratio of downstream to upstream concentration in percent] of less than 0.05% at rated flow... *Filters that fail to satisfy this condition should be* replaced...." (USNRC 1978). Nuclear Regulatory Commission RG 1.140 contains essentially the same information (USNRC 1979). It is recognized that this original guidance should be changed and as indicated earlier, revisions to RG 1.52 are planned. Specifically, Bellamy (1991) notes that guidance in the revised version of the RG will be supplemented such that the 18-month criteria will include "... or once per refueling outage. The requirement for testing after painting, fire, or chemical release will include additional guidance to indicate that this testing need be done only if communication with the system occurred in such a manner that the HEPA filters or carbon adsorbers could become adversely affected by the fumes, chemicals, or foreign materials. Testing will also specifically be required (1) after each partial or complete replacement of a HEPA filter bank. . . ." (Bellamy 1991).

The surveillance leak test of the installed filters should not be confused with the efficiency tests for new individual filters that were described earlier in the report. A polydisperse DOP aerosol is used for the former, in-place test while a 0.3μm monodisperse aerosol is used for the latter. It should be noted that both RG 1.52 and RG 1.140 state that a filtration system satisfying the above 0.05% maximum penetration requirement ". . . can be considered to warrant a 99% removal efficiency for particulates . . ." (USNRC 1978, 1979).

6.2 Adsorbers

The information concerning visual inspection and the air flow distribution and aerosol mixing tests discussed briefly in the previous section also pertains to adsorbers. In the case of the installed adsorber bank, the surveillance leak test is based on a halide challenge gas. The slightly adsorbable and readily desorbed halide (halogenated hydrocarbon refrigerant, fluorocarbon) gas is injected upstream of the adsorber bank. As in the case of the filter test, concentration is then measured upstream and downstream of the bank and percent of penetration determined from the ratio of downstream to upstream values. The recommended test frequency given in ASME N510 (ASME 1989b) is the same as that for the filters and includes acceptance testing, after each adsorber replacement, and at least once each operating cycle. Regulatory Guides 1.52 and 1.140 require testing initially, at intervals of 18 months thereafter, following removal of samples for laboratory tests (if the integrity of the adsorber section is affected), and following painting, fire, or chemical release in any ventilation zone communicating with the system (USNRC 1978, 1979). Both RG also indicate that bypass leakage through the adsorber should be less than 0.05%.

The above planned revisions to RG 1.52 to supplement the 18-month test frequency and require testing only if communication occurred in a manner that components could become adversely affected by fumes, etc., will also influence adsorber testing. In addition, "testing will also specifically be required (1) after each partial or complete replacement . . . of a carbon adsorber in an adsorber section or bank, (2) following detection of, or evidence of, penetration or intrusion of water or other foreign material into any portion of an ESF atmosphere cleanup system, and (3) for adsorber banks following removal of an adsorber sample for laboratory testing if the integrity of the adsorber section is affected" (Bellamy 1991).

As in the case of the polydisperse DOP test of filters, the in-place halide challenge test is a leak rather than an efficiency test. For adsorbers, however, the leak test is supplemented with laboratory tests of aged or used carbon samples to determine system efficiency and remaining capacity for methyl iodide. It is stated in ASME N509 that "sufficient test canisters or other means of obtaining samples . . . of used adsorbent shall be installed in the adsorber system to provide a representative determination of the response of the adsorbent to the service environment over the predicted life of the adsorbent. Test canisters shall be installed in a location where they will be exposed to the same air flow conditions as the adsorbent in the system, shall have the same adsorbent bed-depth as the adsorbent in the system, and shall be filled with representative adsorbent from the same batch of adsorbent as that of the system" (ASME 1989a). Recommendations concerning the number of canisters to be provided based on the expected frequency of operation are also included in ASTM N509. Details concerning the design basis for samplers and the general types are presented in Appendix A of ASTM N509, "Sampling of Installed Adsorbents for Surveillance Testing."

Recommended frequency for the above laboratory adsorbent tests, presented in Table 1 of ASME N510, includes acceptance tests, before each adsorber replacement, and at least once each operating cycle. Supplemental footnotes state that "adsorbents must be tested before installation or replacement to establish efficiency. Samples for laboratory testing should be taken before the routine in-place testing of the installed system to verify the condition of the adsorbent. . . Laboratory tests shall be made to confirm performance at intervals not exceeding 720 hr of system operation or for any system immediately following inadvertent exposure to solvents, paints, or other organic fumes or vapors that could degrade performance of the adsorbent. The

720 hr requirement may be modified based on laboratory test history" (ASME 1989b). As in the case of the qualification of new carbons, the test method and laboratory apparatus described in ASTM D3803 are specified for use in the quantitative evaluation of the retention properties of the aged carbon samples (ASTM 1989). Information to improve repeatability and accuracy is provided in Appendix B of ASME N510, "Additional Guidance for use of ASTM D3803, 1979."

Frequency guidelines listed in NRC RG 1.52 and RG 1.140 for the surveillance test involving adsorbent sampling and laboratory testing are essentially the same as those recommended above. Used carbon decontamination efficiencies are specified for both ESF and normal systems for a variety of air-filtration system locations, carbon bed depths, and relative humidities. For example, a 95% decontamination efficiency is specified for both elemental iodine and organic iodide, for a 50.8-mm (2-in.) bed depth, an ESF system designed to operate outside the primary containment, and where RH is controlled to 70% (USNRC 1978).

7 Conclusions

This section presents conclusions and describes concerns originating from the study. Opinions and recommendations concerning the need for expanded or more comprehensive assessments are also presented. Considerations related to the need for additional studies include the safety significance of failure modes and experience, evaluation of ISMM, and the range of conditions that HEPA filters and adsorbers may experience. Safety significance was emphasized as part of the background information provided earlier in this report. In review, component failure can impact both plant and public safety. Satisfactory performance is essential to ensure control room habitability, and HEPA filters and adsorbers can be the last barrier in preventing the release of radioactivity. Monitoring and surveillance tests have been established to observe HEPA filter pressure drop buildup, check both HEPA filters and adsorbers for bypass or pathways through which air can escape treatment, and determine the retention effectiveness of aged carbon. Conditions range from those associated with normal operating environments to those estimated for reactor accidents. Further development of the preceding considerations, in terms of normal, error-induced, and accident conditions, follow. The need for the expansion of the Phase I interim aging assessment to other air-treatment system components is also discussed.

7.1 Normal Operating and Error-Induced Conditions

Normal operating conditions are characterized as ranging from the exposure of components to dry air containing trace atmospheric contaminants or stressors (e.g., sulfur dioxide and nitric oxides) to relatively humid air and even occasionally to potentially polluted air flows from routine activities such as maintenance and testing. Because of large air flows, even traces of pollutants or contaminants can have a significant cumulative effect on adsorber performance. Two examples of error-induced, degraded adsorber retention efficiency from relatively high contaminant levels (i.e., from the exposure to fumes from cleanup and/or painting) are reported in Sections 5.1 and 5.3. The surveillance test involving the laboratory quantification of the extent of degradation of used carbon samples is specifically designed to reveal aging or premature aging as a result of the normal or error-induced exposure of adsorbers to contaminants or pollutants.

Treatment of large volumes of air containing small particle concentrations can, of course, result in a gradual but continuous increase in pressure drop through filters. Pressure drop or resistance to air flow across HEPA filter banks is monitored. First (1991) notes that "although HEPA filters are qualified to maintain their integrity and filtering efficiency up to a minimum resistance of 2.5 kPa, they are seldom operated up to that resistance level because of fan and fan motor limitations. An increase to 1.0 kPa before change is common for nuclear installations to maintain a large reserve capacity at all times in readiness for extended service life in the event of an emergency" (First 1991). The surveillance test involving the leak test of HEPA filter banks should detect defects in the filter media or unit arising from normal service conditions and that result in bypass, including those that could result from the aging of gasket material. However, neither pressure drop monitoring nor the in-place surveillance test using a polydisperse DOP aerosol will provide indications of aging in terms of reduced filter medium strength. The latter, the lack of indication of reduced strength, may be important when considering reactor accident conditions.

7.2 Reactor Accident Conditions

Nuclear Regulatory Commission regulations require atmospheric cleanup systems to mitigate the consequences of postulated accidents by removing radioactive materials released during the accident from containment vessel or building atmospheres. General Design Criterion (GDC) 41, "Containment Atmosphere Cleanup" of Appendix A, "General Design Criteria for Nuclear Power Plants," to Title 10, Part 50 of the Code of Federal Regulations states that "systems to control fission products . . . which may be released into the reactor containment shall be provided as necessary to reduce, consistent with the functioning of other associated systems, the concentration and quality of fission products released to the environment following postulated accidents . . ." General Design Criterion 61, "Fuel Storage and Handling and Radioactivity Control" states that "the fuel storage and handling, radioactive waste, and other systems which may contain radioactivity shall be designed to assure adequate safety under normal and postulated accident conditions. These systems shall be designed . . . with appropriate containment, confinement, and filtering systems, . . ." General Design Criterion 19, "Control Room" requires that" . . . adequate radiation protection shall be provided to permit access and occupancy of the control room under accident conditions . . . ." (10 CFR 50 1992).

Nuclear Regulatory Commission RG 1.52 presents acceptable methods for implementing the above (and other) GDCs and includes guidance that aging as well as postulated accident conditions should be considered during component design. As indicated earlier, this RG applies ". . . to postaccident engineered-safety-feature atmosphere cleanup systems designed to mitigate the consequences of postulated accidents. It addresses the ESF atmosphere cleanup system, including the various components and ductwork, in the postulated DBA [design-basis accident] environment. . . . systems that must operate under postulated DBA conditions inside the primary containment . . . (i.e., recirculating systems) are designated as . . . [primary]. ESF systems required to operate under conditions that are generally less severe... (i.e., recirculating or once-through systems) are designated as . . . [secondary]. Secondary systems typically include the standby gas treatment system [SGTS] and emergency ACS for the fuel handling building, control room, and shield building. . . . Unless the applicable engineered-safety-feature atmosphere cleanup system operates continuously during all times that a DBA can be postulated to occur, the system should be automatically activated upon the occurrence of a DBA . . ." (USNRC 1978). Information is included in the RG in recognition of the fact that environmental conditions preceding a postulated DBA may contribute to aging and degraded performance of filters and adsorbers. It is specifically noted that aging needs to be considered during design and operation. Furthermore, remembering that the guide was developed for accident mitigation systems, it is also indicated that all components need to be designed for reliable performance in potentially hostile environments.

Typical DBA environmental conditions, including pressure surge, maximum pressure, maximum temperature and RH of the influent, average radiation levels for airborne materials and iodine buildup on the adsorber, and airborne iodine concentrations for elemental iodine and for methyl iodide and particulate iodine, are tabulated in RG 1.52 for the primary and secondary ESF systems described in the preceding paragraph (USNRC 1978). The radiation levels are based on U.S. Atomic Energy Commission (USAEC) RG 1.3 and RG 1.4, which provide acceptable assumptions for use in evaluating the radiological consequences of one of the postulated accidents for boiling- and pressurized-water reactors (i.e., the design basis loss-of-coolant accident [LOCA]). In both of these regulations, it is stated that "twenty-five percent of the equilibrium radioactive iodine inventory developed from maximum full power operation of the core should be assumed to be immediately available for leakage from the primary reactor containment. Ninety-one percent of this 25 percent is to be assumed to be in the form of elemental iodine, 5 percent of this 25 percent in the form of particulate iodine, and 4 percent of this 25 percent in the form of organic iodides" (USAEC 1974a,b).

One of the two major concerns arising from this Phase I study is that the conditions developed for the above design basis assessments may not realistically represent those under which HEPA filters and adsorbers, including aged components, may operate. Design-basis accident evaluations are based on non-mechanistic hypothetical events. Pasedag, Blond, and Jankowski (1981) note that" . . . the Design-Basis Accidents (DBAs) are a set of accidents which have been chosen to envelope the anticipated worst credible conditions in what was perceived to be a very conservative manner. Thus these accidents are not representative of expected or realistic conditions but have been judged to bound any credible accident . . . The DBA-LOCA cannot be expected to be predictive of any specific accident situation." Over the past 10 to 15 years, a large amount of information has been developed that may permit significantly better estimates of the conditions, including radiation levels, temperatures, humidities, and the chemical and physical properties of the materials that aged components might encounter during severe accidents. This information was developed as part of, or in support of, probabilistic risk studies and extensive efforts to reassess source terms (characterization of radionuclide releases from reactor accidents) and has included efforts to provide improved analytical models to describe thermal-hydraulic behavior and mass transport. The latter includes detailed models that take into account the chemical and physical processes associated with the release of fission products from the degraded core and the subsequent transport and retention of material through and in the reactor vessel, coolant systems, containment, and exterior compartments.

A system of computer codes, the Source Term Code Package (STCP), has been developed for analyzing specific postulated accident situations and calculating the quantity, timing, and characteristics of the release of radioactive material to the environment following the incident (Gieseke et al. 1986). This code system or the more recently developed MELCOR computer code for source term and risk assessment analyses could be used to provide significantly more realistic insights into the impact of an accident on filters and adsorbers (Summers et al. 1991). For example, rather than using assumptions, information could be developed from calculations that would be extremely useful in estimating the range and combination of accident conditions that may be encountered, e.g., in providing best-estimates of the likelihood for large particle concentrations and/or moisture conditions that could saturate and overwhelm HEPA filters. An improved definition of accident conditions may also reveal that aging concerns associated with the filters and adsorbers of "normal" treatment systems may equal those of ESF systems and that the designations of primary and secondary for the latter may be trivial in terms of accident considerations. One of the findings resulting from investigations into the TMI accident was the fact that" . . . a nonengineered safety feature filter system designed for normal operation only, i.e., the auxiliary building exhaust ventilation filtration system, greatly reduced the quantity of radioiodine release to the environment . . . The safety grade versus nonsafety grade designation [for the two air treatment systems operating at the time of the accident] was meaningless. . . ." (Rogovin and Frampton 1980).

It should also be noted that in a recent study Beahm, Weber, and Kress (1991) used the STCP to address the issue of the chemical form of iodine in the coolant system and in containment. Using calculated data from seven severe accident sequences, it was found that "in most of the calculations for the seven sequences, iodine entering containment from the reactor coolant system was almost entirely in the form of CsI with very small contributions of I or HI." The study did not address ultimate disposition. Iodine could, of course, subsequently be converted to the volatile form, e.g., if the CsI dissolved in low pH water, was ultimately converted to organic form, or decomposed after being captured on filters. However, the study suggests the perhaps unexpected conclusion that, during accidents, intact HEPA filters could be significantly involved in iodine retention. Furthermore, past design basis evaluations based on the assumption that iodine is primarily in the volatile form may neither be correct nor conservative when evaluating air-cleaning system performance. "Ill-defined accident conditions even where a DBA philosophy is applied" are described as a major problem by the group of NEA experts. As further support for the possibility of nonconservative assumptions, this group also notes that ". . . the DBA may not represent the most severe conditions under which the ACS has to operate" (OECD 1984).

The second major concern arising from the Phase I study is that even if details concerning accident conditions were available, there may be insufficient information to develop reliable predictions concerning the performance of aged components in such potentially extreme environments. In 1984, the group of NEA experts characterized the extent of knowledge that is available to evaluate aged HEPA filter and adsorber performance during accidents as non-existent and for the most part, poor, respectively. The group of experts stated that "...the available information on HEPA filters relates primarily to new filters and virtually no data exist for the normal or accident condition behavior of aged filters. Many ACS HEPA filters remain in place for more than 5 years. It is unrealistic to expect that accidents will occur only with freshly installed HEPA filter banks" (OECD 1984). When considering aged HEPA filters in conjunction with accident environments, the group described the extent of knowledge as non-existent for the following parameters: pressure differential, vibration, high humidity/free water, chemicals, radiation, temperature, and loading capacity. When appraising the extent of knowledge for aged adsorbers, pressure differential was identified as good; vibration as non-existent; radiation as fair; and high humidity/free water, chemicals (weathering), temperature, and loading capacity as poor.

The above relative gloomy picture has brightened somewhat in recent years, presumably as the result of the emphasis on severe accidents. Information has been obtained to at least partially fill in some of the gaps associated with the above parameters. As indicated earlier, above saturation, new or clean filters are vulnerable to deterioration and failure. Obviously, high humidity air flows can jeopardize dirty, particle-loaded filters. The modes and mechanisms of the failure of dust-loaded filters under high humidity conditions are discussed in Ricketts, Ruedinger, and Wilhelm (1987) and in Ruedinger, Ricketts, and Wilhelm (1985). The structural limits, in terms of ΔP at failure, are tabulated in the latter reference for HEPA filters tested under high air humidities at 1700 m^3/h and 50°C. Norman (1987) reports results from the exposure of filter media to steam. The media were dried subsequent to exposure and evaluated with respect to penetration and tensile strength. Increased penetration (using DOP aerosols) was observed upon exposure of the media to high humidities and elevated temperature. Tensile strength decreased to about 40% of its original value. Aspects of the increase in pressure drop of HEPA filters under fog conditions was also investigated by Ricketts, Ruedinger, and Wilhelm (1990). "Airstreams with between 80% and 100% RH primarily threaten dust-loaded filters. At conditions above saturation, new clean filters also become subject to rapid deteriorations in performance." Penetration of water through filters by seepage was characterized as "rather rapid." Such penetration suggests that water soluble salts containing radionuclides could penetrate even intact filters. The performance of downstream adsorption units could also be affected.

Performance limitations and efficiency of and releases from HEPA filters at high temperatures were the topics of three of the papers of Session 3B, "Response of HEPA Filters to Physical Stress," of the 20th DOE/NRC Nuclear Air Cleaning Conference (Kratzke 1989). Effects of temperature on HEPA filter media was again a topic for discussion at the most recent, 22nd, conference where" . . . observed changes in strength and paper stiffness . . . [were] explained in terms of alterations to the binder due to thermal degradation" (Hamblin and Goodchild 1992).

A comprehensive study of the performance of TEDA and KI impregnated charcoals under "reactor accident conditions" is reported in Wren et al. (1989), "The efficiency of these charcoals in removing CH_3I and I_2 was studied as a function of temperature (25° to 80°C), RH (0 to 90% RH), radiation field (0 to 2kGy.hr^{-1}) with and without H_2, contaminants (SO_2, NO_2, NH_3, and MEK) and other factors." Deitz (1985) reports that "based on the experimental results, it appears that reactor-grade activated charcoal can satisfy the stipulation in RG 1.52 that requires successful trapping of radioiodine during a DBA with radiation levels up to [10^7 Gy] 10^9 rads." In fact, it was found that exposure to levels of 10^7 to 10^9 regenerates the iodine isotope exchange capacity. Paraphrasing Deitz, where the trapping of methyl iodide-131 is concerned, radiation improvement rather than damage is observed.

7.3 Conclusions and Recommendations

The Phase I study found that the HEPA filters and adsorbers are considered to have a long service life, especially the filters. Thus, if a severe accident happens it is likely to occur at a time when these two final confinement barriers have been in use for an extended period, even years. Even with existing inspection, monitoring and monitoring methods, aged, and possibly degraded components, could fail to provide the radiation protection needed for safe shutdown or be the weak link that allows the release of radionuclides to the environment. Furthermore, the assessment has revealed the need for an improved definition of accident conditions and the possible need for additional information to evaluate the performance of aged components under such conditions. It is recognized that this improved definition of accident conditions is outside the scope of the NPAR program. It is also recognized that comprehensive assessments limited solely to the development of information for performance evaluation could be made. However, for such studies to be definitive, knowledge concerning challenges must ultimately be obtained, as will be discussed below.

Limited information concerning the range of challenges expected for filters and adsorbers during accidents is already available as a result of source term reassessment and reactor risk studies (Silberberg et al. 1986; USNRC 1990). Additional insights concerning accident conditions should also become available as a result of recent regulatory efforts to provide improved source term definitions. As part of several regulatory activities to incorporate severe accident insights into the safety assessment of future plants, the NRC has issued a proposed revision of the reactor accident source terms. The proposed revision is in terms of fission product composition, magnitude, timing, and iodine chemical form, for release into containment (Soffer 1992). Utilization of these revised source terms should provide improved estimates of accident conditions and the associated challenges to filters and adsorbers. For example, evaluation of iodine chemistry during the portion of the transient that follows release into containment will be needed, resulting in better estimates of whether iodine remains primarily in the particulate form or is converted to a volatile species before it reaches filters and adsorbers.

As previously indicated, even if details concerning accident conditions were available, there may be insufficient information to develop reliable predictions concerning the performance of aged components. Quantification of the structural limits and failure mechanism of aged filters in moist airflows and of the particle loadings that would cause failure of reduced strength and/or particle-laden filter media are examples of potential information needs. Statistically-designed engineering scale experiments, involving aged components and utilizing the improved estimates of the ranges and combinations of challenges, may ultimately be justified. It is also possible that improved estimates of conditions could negate the need for further work, because either moderate or extremely severe stressors are predicted. In the case of moderate stressors, for example, additional study appears unwarranted if near normal operating conditions are identified. In the case of severe conditions, HEPA filters are simply not sized to handle the massive loading of non-radioactive particles that conceivably could result from molten core-concrete interactions.

An improved understanding of severe accident conditions could also further emphasize the importance of other air-treatment system components. For example, along with HEPA filters, greater emphasis would be placed on coarse or roughing pre-filters and even demisters if it were determined that high particle concentrations were involved. As a result, expansion of the Phase I interim aging assessment to other components of nuclear air-treatment systems, namely demisters, heaters, coarse filters, fans or blowers, and dampers is recommended. Several of these components, although not designed to retain radionuclides, could mitigate the impact of conditions that threaten to cause failure of aged HEPA filters and adsorbers during accidents. The aging of these "protective" components is, therefore, also of concern. The information that is developed for fans can be used in the assessment of other ESF systems specifically installed to ensure public and plant safety. In addition to air cleaning, fans or blowers are included in heating, ventilation, and air conditioning and containment cooling and pressure suppression systems.

Finally, it should be emphasized that information used to estimate the performance of aged air-treatment system components can have a significant impact on the calculated consequences of postulated accidents. This is illustrated by the analyses of a postulated loss of decay heat removal accident at Browns Ferry. The SGTS failure model used, listed as the area of greatest uncertainty with respect to informal sensitivity analyses, includes best estimates concerning the filter loading that will cause failure, the type of failure, and the functioning of the adsorber under the projected accident environment. It is noted that "the SGTS failure model assumes prime importance because the SGTS is the last barrier to the atmosphere in this accident sequence" (Wichner et al. 1984).

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10. SUPPLEMENTARY NOTES

11. ABSTRACT *(200 words or less)*

A Phase I aging assessment of high-efficiency particulate air (HEPA) filters and activated carbon gas adsorption units (adsorbers) was performed by the Pacific Northwest Laboratory (PNL) as part of the U.S. Nuclear Regulatory Commission's (NRC) Nuclear Plant Aging Research (NPAR) Program. Information concerning design features; failure experience; aging mechanisms, effects, and stressors; and surveillance and monitoring methods for these key air-treatment system components was compiled. Over 1100 failures, or 12 percent of the filter installations, were reported as part of a Department of Energy (DOE) survey. Investigators from other national laboratories have suggested that aging effects could have contributed to over 80 percent of these failures. Tensile strength tests on aged filter media specimens indicated a decrease in strength. Filter aging mechanisms range from those associated with particle loading to reactions that alter properties of sealants and gaskets. Low radioiodine decontamination factors associated with the Three Mile Island (TMI) accident were attributed to the premature aging of the carbon in the adsorbers. Mechanisms that can lead to impaired adsorber performance include oxidation as well as the loss of potentially available active sites as a result of the adsorption of pollutants. Stressors include heat, moisture, radiation, and airborne particles and contaminants.

12. KEY WORDS/DESCRIPTORS *(List words or phrases that will assist researchers in locating the report.)*

HEPA filters
Phase I aging
adsorbers
air-treatment system
design feature
failure experience
aging mechanisms
stressors

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