Fission Product Removal in Engineered Safety Feature (ESF) Systems

Data Base Assessment and Suggested Experimental Program

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ABSTRACT

The available data base on the fission product removal capabilities of nuclear reactor Engineered Safety Feature (ESF) systems was reviewed and assessed. The systems considered included pressure suppression pools, ice condenser systems, containment sprays, filter systems and containment air coolers. Based on this assessment, a research program was recommended to expand this data base to support the development of mechanistic models and computer codes for the prediction of ESF system fission product removal. This research program included experimental efforts to better define the performance of ice condenser systems, expand the range of data available on water spray systems and to investigate the behavior of containment air coolers, demisters and fans in the presence of aerosols typical of those expected following a severe accident.

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SUMMARY

This report reviews the currently available information on the removal of fission product aerosols by Engineered Safety Feature (ESF) systems. The ESF systems that received principal attention were:

- Pressure suppression pools
- Ice condenser systems
- Containment sprays
- Filter systems
- Containment air coolers.

The nature and status of planned and ongoing research in the area of fission product removal by such systems was also reviewed. Based on these reviews, the suitability of the existing and potential future data base to support the development and verification of mechanistic models was assessed. This assessment revealed that this data base may be adequate in some areas but not in others. Areas where additional information may be desirable include:

- The performance of ice condenser systems
- Aerosol removal by sprays
- Plugging of containment spray (CS) pump inlet screens by debris resulting from a severe accident
- Deposition of aerosol materials in containment coolers
- Operation of containment filter system components following a severe accident.

A recommended research program was developed that would address these areas. Major experimental activities included in this program are:

- An experimental study of fission product scrubbing in ice condenser pressure suppression systems
- An extension of the earlier CSE containment spray tests to a wider range of variables
- Studies of the interaction of aerosols with demisters and air cooler coils
- An investigation of the operation of fans and motors of the types used in ESF systems in high temperatures and dense aerosols expected following a severe accident.

In addition to these experimental studies, several engineering studies and surveillance activities were recommended to help define and guide future analytic and experimental activities regarding the interactions of aeroso!s with ESF systems. .

CONTENTS

ABST	RACT.		• • • • • • • • • • • • • • • • • • • •	iii
SUMM/	ARY			v
1.0	INTRO	DUCTION	1	1.1
2.0-	EXIST	ING DAT	A BASE	2.1
	2.1	OVERVIE	W OF SOURCE TERM DATA BASE	2.1
		2.1.1	Fission Product Release from the Fuel	2.2
		2.1.2	Formation of Aerosols	2.4
		2.1.3	Transport and Removal of Fission Products by Natural Processes	2.5
		2.1.4	Release Paths to the Environment	2.7
	2.2	ESF SYS	STEMS FISSION PRODUCT REMOVAL	2.8
		2.2.1	Pressure Suppression Pools	2.8
		2.2.2	Ice Condenser Systems	2.12
		2.2.3	Containment Sprays	2.13
		2.2.4	Pumping Systems	2.14
		2.2.5	Filter Systems	2.15
	2.3	ASSESS	MENT OF THE ESF SYSTEMS DATA BASE	2.17
3.0	ADDI	TIONAL D	DATA BASE REQUIREMENTS	3.1
	3.1	SUPPRES	SSION POOLS	3.1
		3.1.1	Models for Predicting the Performance of Suppression Pools	3.1
		3.1.2	Data Requirements for Suppression Pools	3.2
	3.2	ICE BEI	DS	3.3
		3.2.1	Models for Predicting the Performance of Ice Beds	3.3
		3.2.2	Data Requirements for Ice Beds	3.3
	3.3	CONTAIN	MENT SPRAYS	3.4
		3.3.1	Models for Predicting the Performance of Containment Sprays	3.4
		3.3.2	Data Requirements for Containment Sprays	3.4
	3.4	FILTER	SYSTEMS	3.5
		3.4.1	Filter System Analysis Methods	3.5
		3.4.2	Data Requirements for Filter System Performance	3.5
	3.5	CONTAI	NMENT COOLERS	3.6

		3.5.1	Models for Performance of Containment Coolers 3.	, 6
		3.5.2	Data Requirements for Containment Cooler Performance 3.	.6
4.0	RECON	MENDED	RESEARCH PROGRAM 4.	. 1
	4.1	SUPPRES	SSION POOL STUDIES 4.	.1
	4.2	FISSIO SUSPRE	N PRODUCT SCRUBBING IN ICE CONDENSER PRESSURE SSION SYSTEMS	.2
	4.3	SCRUBB	ING OF FISSION PRODUCT BY WATER SPRAYS	. 2
		4.3.1	Containment Spray Inlet Screen Plugging 4.	.3
	4.4	FILTER,	/COOLING SYSTEMS 4.	3
		4.4.1	Aerosol Removal by Containment Air Coolers 4.	,4
		4.4.2	Influence of High-Density Aerosols on Fan/Motor Performance4.	.5
		4.4.3	Aerosol Removal by Demisters 4.	. 5
5.0	PEER	REVIEW	5.	.1
	5.1	THE PE	ER REVIEW GROUP	.1
	5.2	RESULT	S OF THE PEER REVIEW 5.	.1
6.0	REFE	RENCES.		.1

FISSION PRODUCT REMOVAL IN ESF SYSTEMS: DATA BASE ASSESSMENT AND SUGGESTED EXPERIMENTAL PROGRAM

1.0 INTRODUCTION

Following the TMI-2 incident in 1979, regulatory interest in severe accidents involving core damage and melting has intensified. Particular attention has been directed towards factors which contribute to the determination of the source terms for various accident sequences. One factor receiving attention is the Engineered Safety Feature (ESF) systems and their potential role in mitigating fission product releases following these types of accidents. ESF systems of particular interest with regard to release mitigation include: 1) spray systems, 2) ice condensers, 3) pressure suppression pools, 4) filter systems, and 5) air coolers. Also, several "add-on" systems have been proposed, including vent-filter systems, core retention systems and hydrogen control systems; however, these have not been used in commercial power reactors.

These ESF systems can influence the fission products in the containment volume following a severe accident in two ways:

- 1) They influence the time-sequence of events by providing cooling before, during, and after fuel melting and vessel melt-through.
- They can directly remove a substantial portion of the fission product aerosols.

At the request of the U. S. Nuclear Regulatory Commission the Pacific Northwest Laboratory has completed a study with the following objectives: 1) to review and assess the experimental data base for the prediction of the performance of ESF systems in the removal of fission product aerosols from the containment atmosphere, and 2) to formulate a recommended experimental research program to support analytic efforts to include realistic ESF systems behavior in methodologies for predicting the source terms for releases from light water reactors (LWRs). The Pacific Northwest Laboratory (PNL) is operated for the U.S. Department of Energy by Battelle Memorial Institute.

Section 2.0 of this report reviews the existing data base. Section 3 discusses additional data base requirements of the five ESF systems listed above. A recommended research program for fulfilling these additional data requirements is presented in Section 4.0. Section 5.0 reports the results of a review of this document by a separate group of researchers working in the areas of the source term and fission product removal performance of ESF systems.

2.0 EXISTING DATA BASE

The performance of ESF systems in removing fission products from a reactor containment system following a severe accident that involves core damage and melting is only one factor that must be addressed in the determination of the source term. Other factors include:

- the time sequence of events in the accident event,
- the time-temperature history of the fuel and the melt,
- extent of damage to the core,
- thermodynamic, hydrodynamic and chemical conditions in the vessel and containment
- fission product release phenomena
- aerosol formation
- fission product transport in the vessel and containment, including natural depletion mechanisms
- pathways to the environment.

This section provides, as background, an overview of the general status of the data base for definition of the source terms for light water reactors. Section 2.2 presents a review of the available data base on the performance of ESF systems in removing fission products. Section 2.3 is devoted to an assessment of this data base.

There is not general agreement regarding all aspects of fission product release, transport, and deposition following a severe accident. In the following paragraphs, some of the more important data and hypotheses regarding these processes will be presented. Because this section is intended as a brief overview only, the information provided is not necessarily complete, nor should it be considered as endorsing any one point of view over another.

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2.1 OVERVIEW OF SOURCE TERM DATA BASE

The fission product release during a severe accident involves a complex sequence of events and is strongly dependent on the temperature history of the fuel and the thermal and hydraulic conditions in the vessel and containment system. Prior to the TMI-2 incident, only limited work was done to analyze these events and conditions. Since that time, a number of computer codes have been developed to study severe accidents involving core damage and melting, including MARCH (Battelle), INCOR (Electric Power Research Institute-EPRI) and KESS (Kraftwerk Union AG). These codes are relatively sophisticated in the evaluation of events below 2200°F where the data base on $Zr/fuel/H_{20}$ interactions is extensive. Also, they are relatively sophisticated in their descriptions of steam condensation and structural response aspects. However, events

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at temperatures above 2200°F are approached only parametrically because few data are available. Other codes including INTER, CORCON (Sandia National Laboratories - Sandia), GROWS-II (Argonne National Laboratory-ANL) and WECHSL (Karlsruhe) have been developed for the study of the molten core/concrete interactions.

Little large-scale experimental work on fuel melting and fuel slump has been performed to verify these codes. Most planned experiments generally involve only single fuel rods or small bundles of fuel rods; e.g., the PBF tests will use only a 32-rod bundle, 0.9 m long. The largest test being planned is the LOFT L2-6 experiment. However, this test will be the last test of the LOFT Program since it will make LOFT unusable for further experimentation, and the schedule for this test is unclear at this time. Therefore, it appears that suitable large-scale data for code verification will not be available in the near future. Large-scale experiments are being performed at Sandia, ANL and Germany on the interaction between corium, a nonradioactive molten core simulant and concrete. These experiments are being used to further develop and verify codes that describe events following reactor vessel melt-through.

2.1.1 Fission Product Release From Fuel

The progression of fuel damage with increasing temperature was described by Chung and coworkers (1981) as follows:

(<u>></u> 700°C)	-	Ballooning of Zircaloy cladding
(<u>></u> 750-1050°C)	- - -	Rupture of Zircaloy cladding Oxidation of metal components/hydrogen generation Embrittlement by oxidation
(1400-1900°C)	-	Reaction between solid UO_2 and metallic Zircaloy
(>1980°C)	- - -	Melting of remaining Zircaloy Breach of ZrO ₂ shell by liquid UO ₂ Flow-down of liquified fuel and Zircaloy
(~2700°C)	-	Melting of remaining solid ZrO ₂
(~-2820°C)	-	Melting of remaining UO2

The possible role of melting of control rod materials (Cd, In, and Ag alloys at $\sim 800^{\circ}$ C) is not completely understood. The core may be reflooded and uncovered repeatedly during the above sequence events, causing additional fragmentation of fuel components by thermal shock.

Lorenz described the mechanisms that control the release of fission products during the core disintegration (Lorenz et al. 1978 and 1974; NUREG/CR-0091; NUREG/CR-1288). These are:

1) Burst Release

This burst release occurs when the overheated fuel rod cladding ruptures (750 to 1050°C) and releases the entire inventory of noble gases from the plenums and open voids. This can amount to 0.25 to 25 percent of the total stable and long-half-life isotopes of fission gases. Shortly thereafter, gases shallowly embedded in the fuel and cladding will be released, adding an additional 1 to 1.5% of the total fission gas inventory. Cesium (Cs) and iodine (I) will also be released in smaller amounts; for a pressurized water reactor (PWR), this release will be approximately 0.02% and 0.04% of stable Cs and I, respectively.

2) Diffusional Release

After the burst release, the cesium and iodine will slowly diffuse out of the ruptured fuel elements.

3) Grain Boundary Release

At approximately 1350°C, the fission gases, cesium and iodine, accumulated in the grain boundaries will be released. In high burnup fuels, this release would involve up to 20% of the stable isotopes of these elements.

4) Diffusional Release from UO₂ Grains

Following the burst release, the release from the UO_2 grains is insignificant. However, as the fuel heats, the remaining noble gases, Cs and I, are released at a rate which doubles about every 100°C. At 2000°C the release rate is approximately 10%/min.

5) Release from Molten Fuel

The details of the fission product release from molten fuel is not well understood because of the complexity of the chemical transformations of fuel, control rod, and cladding materials which occur simultaneously with the melting process.

There has been considerable recent discussion in literature regarding the chemical form of iodine produced by the degraded fuel. It is now generally accepted that iodine is released from the fuel as cesium iodide (CsI) because of the reducing environment in the core during the release process (Pasedag et al.; Chellew et al., Ritzman et al.; Genco et al.). This species is less volatile than elemental iodine and is soluble in water. This reasoning has been used to account for the smaller than expected iodine releases observed in release incidents.

There also has been some concern regarding tellurium since it can decay to radioactive iodine. However, it appears that this tellurium may not be a particular problem. Work at ORNL has indicated that perhaps no Te is released during fuel melting (Parker et al. 1981). Tellurium tends to combine with Zircaloy cladding to form ZrTe (Genco et al.) and Te_3ZrO_5 (NUREG/CR-0274), which has a strong tendency to plate out on system surfaces (Allison; Allison and Rae).

The current method of calculating the release of fission products is described in NUREG-0772. This method is based on experimental data obtained by Lorenz (NUREG/CR-0274, NUREG/CR-0222; NUREG/CR-1386, NUREG/CR-1773), Parker et al. (1963, 1967), and Albrecht et al. (1979a, 1979b). These experimental data were analyzed to give smoothed curves of iodine, cesium, and tellurium release as functions of time and temperature. It was recognized that the data base was qualitative and inadequate in several areas. However, these data are the best currently available, and investigations to better define the releases of these materials are currently under way.

2.1.2 Formation of Aerosols

If a core melt occurs, aerosols can be produced by condensation of vaporized fission products and other core materials. As these vaporized materials are transported to cooler core regions, they can condense, forming small $(0.01-0.05 \ \mu m)$ particles. Subsequently, other aerosols can be generated during

- reactor vessel melt-through
- oxidation reactions (steam explosions)
- molten core/concrete interactions.

These aerosol particles can grow by agglomeration and by the condensation of water vapor on them. Particles of less than $\sim 10 - \mu m$ aerodynamic mass median diameter (AMMD) will tend to stay airborne for a long time. Larger particles will fall out more rapidly. Thus, aerosol behavior depends strongly on physical processes (and particulate solubility) and is largely independent of the chemical form of the particles (Morewitz).

A number of experimental investigations have been performed to define the AMMD of condensed aerosols of the uranium oxides. These experiments have generally agreed that the AMMD for the oxides of uranium were approximately proportional to the 2.5 root of the released aerosol concentrations at low vapor concentrations (<60 g/m³) (Parker et al. 1967; Baurmash et al.; Parker et al. 1979). In the presence of condensing steam, agglomerated aerosols (which have the appearance of microscopic chain-like structures with many branches) contract into compact clusters because of the effect of the surface tension of water condensing on them.

In a review of recent experiments on fission product source terms for LWR meltdown accidents, Malinauskas et al. concluded that the most probable aerosol particle size was <0.5 μ m at temperatures from 1800°C to 2700°C.

It was noted that the least volatile components were concentrated in the larger particles, while the highly volatile components were concentrated in the smaller particles. At higher concentrations ($\sim 200 \text{ g/m}^3$), Morewitz et al. found that the fallout of an aerosol of UO₂ showed two distinct stages. In the first stage, which lasted approximately 10 seconds, most of the mass precipitated out

of the aerosol. In the second stage, the mass was more persistent. The average projected diameter of the particles during the early stage was approximately 40 μ m. Later, Nelson and Beyak found an AMMD of 39 μ m for UO₂ aerosols at an average concentration of 65 g/m³.

Following the pressure vessel melt-through, the molten core/structural material mixture will come into contact with the concrete floor of the containment. Experiments have been performed to investigate the subsequent aggressive interaction of the molten material with the concrete (Powers and Arellano; Powers et al.). Upon contact, hydrates and carbonates are thermally decomposed to steam and carbon dioxide. The concrete is rapidly eroded, producing substantial amounts of noncondensible gases. These gases sparge through the melt, forming aerosols of $1-\mu m$ typical mean size by two mechanisms:

- 1) Vaporization and subsequent condensation of volatile species
- Mechanical agitation of the melt by sparging gases--These results are being used as a basis for computer models describing this aerosol production mechanism.

In general, it can be concluded that the available data on fission product release is quite extensive. However, more information is required in the area of rate processes that pertain to the progression of severe accidents in LWRs. Additional information is needed to investigate the effects of sample size, pressure, and gas flow rate and to verify rate data obtained with simulants. Rate data are especially needed for structural material aerosol releases resulting from molten core/stainless steel and concrete reactions. Programs addressing these needs are now under way in the U.S. (Oak Ridge National Laboratory-ORNL, ANL, Sandia, and Idaho National Engineering Laboratory-INEL) and Germany (KFK/PNS). However, these programs appear to involve measurements at 1 to 2 bars at relatively rapid bulk flow rates. More data is required at 100 to 150 bars at relatively slow volumetric flow rates to address other important accident situations identified in recent Probabilistic Risk Assessment (PRA) studies.

2.1.3 Transport and Removal of Fission Products by Natural Processes

Natural processes occurring in the reactor core and the containment system have a strong influence on the attenuation of fission products following a severe accident and, therefore, on the fission product/carrier gas composition as it reacts with the ESF systems. The principal natural processes include

- plate-out on core, cooling system and containment surfaces
- dissolution and entrapment by water
- agglomeration and settling.

According to experimental work at ORNL, nearly all of the I, Te and Cs and half of the other fission products are released as the fuel is melted. All of these fission products with the exception of the I, Te and Cs will plate-out in the region around the fuel. The I, Te and Cs combine in the fuel rods (Forsyth et al.; Cubicciotti and Sanecki; NUREG/CR-0722) before release, forming CsI and Cs₂Te. These compounds are water soluble and can be entrapped in any water present in the core or containment system. In a steam environment the Cs₂Te may undergo hydrolysis. Some thermodynamic studies have indicated that H₂Te is the most stable species (Torgerson et al.; Ritzman and Cubicciotti). Tests at General Electric showed that Cs plates out on surfaces at 1000°F to 1800°F and I at 80°F to 600°C (Roberts). In other work, Castlemen found that 90% of the airborne iodine released in steam environment will collect on surfaces below 120°F.

Other experiments on the plate-out behavior of fission products in single and multiple fuel rods were performed at ORNL (Parker et al. 1963). With single pins, only very minor amounts of particulate activity were observed to escape the furnace liner surrounding the rod. Furthermore, with multiple fuel rods, plate-out on the surrounding undamaged fuel rods reduced the release by a factor of 100.

Similar evidence was found at Hanford (Hilliard et al.) in the Containment Systems Experiment (CSE). In these experiments the release of simulated fission products from molten fuel was investigated in helium, air and steam. It was found that natural processes were extremely effective in limiting the amount of fission products leaking from the containment vessel. These processes caused reductions of fission product leakage of 10^{-2} and 10^{-3} for iodine and cesium, respectively, during the initial 24-hour period and 10^{-3} and 10^{-4} during a subsequent 24-hour period. Removal of simulated fission products was observed to occur in the furnace, injection line, containment vessel and the leak paths from the containment to the atmosphere. Two important processes identified in the removal of fission products from the containment atmosphere were

- Surface deposition of gases and particles by diffusion augmented by condensing steam
- Gravity settling of fission products attached to particles.

Similar results were observed by other investigators (Cottrell et al; Parker).

Experiments at Rockwell International considered the agglomeration and fallout of high-concentration aerosols (Morewitz et al.). Two phases were observed in the fallout behavior: the first lasted for approximately 10 s and accounted for 90% of the removal, and the second persisted for tens of minutes. It was observed that at high concentrations (70 to 100 g/m³) agglomeration is rapid (milliseconds) and the resulting products are large (100 to 400 μ m). These giant agglomerates fall rapidly and sweep out additional aerosol mass as they fall during their gravitational fallout.

Levenson and Rahn summarized some of the more important characteristics of released fission products following a degraded core incident and subsequent natural removal mechanisms:

- Highly concentrated aerosols coalesce rapidly; low-density aerosols increase their effective density rapidly in the presence of water and serve as condensation nuclei.
- Agglomerated aerosols formed at high density are dense and settle out close to their source.
- Iodine in its many forms is chemically and physically reactive and is easily immobilized by reaction with organic coatings (paint) inside the containment vessel.
- The containment system and equipment inside it provide a large surface area for fission product plate-out and adsorption.
- The moisture in the reactor containment building will cause most of the soluble airborne fission products to go into solution.
- The presence of large amounts of water and vapor along with the heat capacity of the containment building would be sufficient to immobilize a large fraction of the radioactivity.

The net effect of these natural mechanisms is to 1) reduce the magnitude of fission product released from the fuel, and 2) reduce iodine and particulates relative to the noble gases.

Most of this knowledge of the behavior of fission products in the primary cooling system inferred from behavior observed in test systems originally constructed for other purposes and is only qualitative. More information is required using systems more closely simulating the thermodynamic conditions expected during degraded core incidents. At present, experiments are under way in Germany and Sweden to better define the mechanisms of fission product removal from the containment under simulated conditions for LWR degraded-core accidents.

2.1.4 Release Paths to the Environment

Release pathways to the environment are a matter of central importance in the definition of the source terms. These pathways, which are very specific to both the plant and the hypothesized accident sequence, can be grouped in five broad categories (Lavine et al.):

- 1) Energetic rupture due to explosive events such as steam explosions and hydrogen detonations
- 2) Rupture from overpressurization from steam production, the generation of non-condensible gases, or hydrogen combustion
- 3) Containment leakage

- 4) Basemat melt-through
- 5) Containment bypass.

Recent studies have indicated that steam explosions may pose little significant threat to the containment (Lavine et al.). However, the "steam spike", a less rapid pressure buildup in the containment, may be very important. Significant ongoing research on hydrogen production and combustion is under way at Sandia, LASL, INEL, AECL, and Battelle-Frankfurt, and an improved understanding of these phenomena and their potential effects on containment integrity is being developed. There appears to be a real need for additional work to: 1) define potential leakage path configurations and 2) define the leak rates and fission product removal rates in these leakage paths.

No tests to destruction have been performed on large-scale containment vessels to identify failure modes and leakage configurations. Although such tests would fulfill an important need, their performance in the foreseeable future is questionable because of the prohibitive costs.

2.2 ESF SYSTEMS FISSION PRODUCT REMOVAL

This section addresses information regarding the capabilities of certain ESF systems to remove fission products gases and aerosols from the containment volumes.

2.2.1 Pressure Suppression Pools

BWR containment systems are arranged so that for most accident sequences, the steam and fission products released from the reactor vessel will be transported to a pressure suppression pool filled with water. In the suppression pool, the steam will be condensed and the fission products will be subject to scrubbing.

Recently, General Electric (GE) reviewed the technical literature on fission product retention by subcooled and saturated pressure suppression pools (Rastler). Applicable data identified by GE are summarized in Table 2.1. Also included in this table are the results of subsequent studies reported by Rastler on the retention of insoluble particulates in water pools. Most of the data summarized in Table 2.1 consider elemental iodine; other data concern the scrubbing of CH_{3I} , HI, HIO and small insoluble particles. There appears to be no data on the removal of CSI.

Analysis of the data reported in Table 2.1 revealed that none were gathered under conditions duplicating a severe accident in a BWR. For example, in some tests the simulated fission products were transported to the pool by means of violently flashing steam from a depressurizing pressure vessel. However, in a BWR degraded core accident, the release of fission products is expected to be a relatively slow and steady process. In other tests, the realism of the data was adversely influenced by shallow pool depths or nonrepresentative particulate size distributions. Based on an analysis of the data summarized in lines 1 through 12 in Table 2.1 General Electric was unable to estimate representative suppression pool decontamination factors (DF). However, lower bound estimates were made and are summarized in Table 2.2.

GE estimated that these DFs could be higher by several orders of magnitude under conditions more representative of a severe accident.

Following a molten core/concrete interaction, Sandia data indicated that the vapor released would condense and agglomerate to form a $\sim 2-\mu m$ aerosol in the cooler regions of the containment (Berman). Based on the Sandia data and on data on the scrubbing of volatile I2 and NiCr particulates summarized in Table 2.1, General Electric estimated DFs of at least 100 or greater for a subcooled pool during a core-concrete interaction event (Rastler). Data to confirm these DF estimates are not currently available.

A more recent General Electric study considered the scrubbing of particulates in single gas bubbles (Marble et al.). Activated Eu, 0.05 to 10 μ m, was the particulate material, and air was the carrier gas. Decontamination factors of 100 to 4200 were measured and found to depend on submergence height (i.e., bubble residence time) and particle size.

These data were used as a basis for a computer model of the particulate scrubbing behavior of a BWR/6 Mark III containment system. This model predicted decontamination factors of 9 x 10^3 and 6 x 10^2 for steel/corium and concrete/ corium, respectively, for discharges through the horizontal vents. For the vent with the GE "X-quencher", DFs of 4 x 10^4 for steel/corium were predicted.

A program considering the removal of radionuclide aerosols in BWR pressure suppression pools and PWR quench tanks is currently under way at Battelle, Columbus Laboratories (BCL) under the sponsorship of the Electric Power Research Institute (EPRI). This program includes scrubbing experiments to allow measurement of the decontamination factors under accident conditions, and hydrodynamic experiments to determine the type of gas/water interface, bubble rise velocity, and the bubble size distribution tests will be performed:

TABLE 2.2. Minimum Decontamination Factors for a Suppression Pool

	Subcooled Pool	Saturated Pool
I ₂	10 ²	30
CSI	10 ³	10 ²
Particulates	10 ²	10 ²

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	Experimenter/ Date	Pool Size (gal)	Depth Injection (ft)	pH	Temp (°C)	Carrier	Flow Rate	Simulated f.p.	DF	Comments
1	GE (1959)	1000	1.5	7	Subcooled	Flashing Water and Air	Rapid Blowdown	Xe, Kr I2 NaI (Soluble) ZnS ₂ (Insoluble) (2 µm)	2-4 10 ⁵ -10 ⁶ 5×10 ⁵ 5×10 ⁷	Facility simulated LOCA Blowdown to Pressure Suppression System
2	Hillary et al. (1966)	2000	2	6.8	10-60	Steam, Air, and Steam, Air Mixtures	Steady Steam: 3-9 lb/sec Air: 0.1-7.0 lb/sec	I2 Ni-Cr (0.06 µm)	14-320 15-1680	Scale Model of a SGHW Reactor
3	Diffey et al. (1965a)	3	.17	-	50	Steam/Air	Steam 4 1b/sec-ft ²	I ₂ (0.6-40 ppm I ₂) I ₂ (0.01-0.4 ppm I ₂)	10-500 10-500	Includes both large and small scale tests
		150	1.7	-	50	Mixtures	Air 3 lb/sec-ft ²	HI CH ₃ I Ni-Cr (0.06 µm)	10-1000 1.5-5 50-100	
4	Dadillion and Geisse, France (1967)	11,000	6-20	-	Subcooled	CO ₂ (400°C and 280 psi)	.02-04 lb/sec	I ₂	70-10 ⁴	PIREE Experiment
5	Stanford and Webster ORNL (1972)	260	4	-	Subcooled	Steam/Air Mixture (Air O-2 wt %) (125 psig sat. steam)	22-66 lb/sec-ft ² Steady	I ₂ (0.5-10 ppm)	70-11000	Simulated pressure sup- pression pool
6	Siegwarth and Siegler GE (1971)	150	1-4	7-10	32-66	Flashing Water and Air	Rapid Blowdown	CH3I	1.1-3.2	Simulated LOCA in 1/10,000 scale model of BWR MK-I
7	Marviken Sweden (1974)	10 ⁵	9	-	20-40	Flashing Water and Air	Rapid Blowdown	CH3I	1.2-4.9	Full scale reactor
8	McGoff and Rodgers MSA (1957)	300	10-20	-	Subcooled	Flashing Water and Air	Rapid Blowdown	I2 Y203 RБ С1	2×10 ⁵ 3×10 ⁵ 10 ⁷	Study of venting in seawater
9	Malinowski et al. <u>W</u> (1971)	30	3-8	4-5	49	Не	0.05 lb/sec-ft ²	I ₂ (20 mg/liter) .	88-1500	Glass column 9 in. OD and 8 ft high
10	Hillfard HEDL (1981)	-	2	-	Subcooled	N2	0.5 ft ³ /min	Na ₂ 0 (4 µm)	20	Scrubbing of particles by water
11	Devell et al. (1967)	530	3-12	0-12	Saturated	Superheated Steady (175-300°C)	Steady 0.1-0.4 kg/sec 90-360 lb/sec-ft ²	I ₂ (0.1-2 ppm)	2-200	Saturated Pool Testing included both laboratory and large scale tests
12	Strikovitch et al. (1964)	-	-	5.5-10	Saturated 120-180	Steam	Steady 0.2-1.0 kg/hr	I ₂ (2.5-250 ppm)	10-250	Small scale laboratory test
13	Marble et al.	45	34-167 cm	-	60	N ₂ , Air 20°C	Single bubbles 0.4-1.4 cm	Eu ₂ 0 ₃ (0.05-10 µm)	100-4000	Single bubble tests

TABLE 2.1. Summary of Pool Scrubbing Tests

- Single-orifice tests: 0.391-in.-ID BWR X and T quencher and 0.75-in.-ID PWR quench tank
- 2) Multiple-orifice tests
- 3) Large-scale injections simulating downcomer and horizontal vents of up to 6-in.-ID.

Model aerosols will be chosen to simulate soluble CsI and insoluble Te.

The experiments will be conducted in two tanks, a clear plastic tank (6 ft OD and 8 ft high) and a steel tank (10 ft OD and 14 ft high) with glass view ports. Both tanks have steam injection, and the steel tank can be operated at T_{sat} . Fission product simulants will be used during the tests. Particle concentration will be measured gravimetrically, and an optical measurement system will be used to measure the particle size distribution.

The range of experimental parameters are:

Particle size range	0.1 - 2.0 pm
Pool temperature	ambient to T _{cat}
Carrier gas	0-100% steam in air
Submergence height	to 14 ft maximum

In Sweden, the Marviken Aerosol Transport Tests (ATT) are under way and will consider, in part, the removal of simulated fission product aerosols in a 30,000-kg quench tank and a 150,000-kg wetwell. The ATT program is being funded by a consortium led by Studsvik Energiteknik AB, Sweden; U.S. participants include NRC and EPRI. This program has two objectives. The primary objective is to determine the transport of relatively dense aerosols through a full-scale reactor system. The second objective is to study the transport of volatile fission products through a full-scale reactor system.

The Marviken reactor was originally built as a boiling heavy water reactor and will be modified to resemble a LWR system. These modifications will simulate the dimensions and surface areas of a U.S. PWR as closely as possible.

The aerosol tests will be performed with a core material simulant, "corium," which consists of a mixture of uranium oxide, zirconium oxide and iron. Approximately 800 kg of corium will be vaporized and heated to 2500°C with a plasma arc heater in a tank representing an LWR reactor vessel. The aerosol will be blown by steam/air from this tank through pipes, a dry compartment and a wet compartment. Samples will be taken at intervals to characterize the aerosol in each volume. These samples will include airborne material, deposited material, and liquids. Measurements will be taken of temperatures, pressures, steam quality and vapor pressure.

The fission product transport tests will be performed by vaporizing a fission product simulant, "fissium", which consists of CsI, CsTe, Sr and Fe. Approximately 100 kg of fissium will be evaporated by raising its temperature in nitrogen and steam from 300°C to 1500°C over a period of 2 hours. Samples of

airborne material and liquids will be obtained at frequent intervals at various locations and analyzed for concentration, particle size, and chemical concentration. The fissium tests will be performed both with and without a corium aerosol.

Other system modifications are being planned to simulate BWR configurations.

Present schedules call for the performance of the initial fissium experiments in 1983 and the corium experiments in 1984. Other experimental details may be obtained from the reference, "Aerosol Transport Tests," and from Collen and Mecham.

2.2.2 Ice Condenser Systems

The ice condenser system is used in some Westinghouse PWRs to suppress the rise in pressure that would result from a rupture in the reactor cooling system. It also would tend to reduce the fission products in the containment air by entrapment and dissolution.

A typical ice condenser system consists of an annular compartment which contains approximately 1.1 million kg of flaked borated ice at the outer circumference of the containment vessel. This annular compartment connects the 7800-m³ compartment containing the reactor and cooling system with the 18,500-m³ upper containment compartment. Following a break in the reactor vessel or cooling system, the discharged steam, fission products, and reactor compartment air will flow through the ice condenser, where the steam will be condensed and fission products attenuated. The circulation of the postaccident containment atmosphere through the ice condenser is maintained by two axial flow fans with approximately 80,000-cfm capacity. These fans transfer air from the upper to the lower compartments, thereby inducing a flow through the ice compartments. Dampers are provided to prevent reverse flow through the fans. The main benefit claimed for the ice condenser is a substantial reduction in the required volume and the pressure capability of the containment vessel (50% less volume and 75% lower pressure) (Liparulo et al.; McCurdy et al.).

Malinkowski performed an experimental study to determine the effects of vapor concentration, ice additives, ice loading, vapor temperature, flow characteristics and iodine concentration on the removal of gaseous elemental iodine from the vapor stream entering an ice condenser unit. Experiments were performed in two separate simulated ice condensers; smallscale tests were performed in an 18-in. by 1.5-in.-ID glass tube, "large"-scale tests used a 4-ft by 9-in,-ID tube. The following results were obtained:

- Alkaline additives enhance the retention of iodine in the ice melt by hydrolysis reactions that convert the iodine to nonvolatile soluble forms of iodide and iodate. Sodium tetraborate $(Na_2B_4O_7 \cdot 10_{H_2}O)$ has a high capacity for iodine removal.
- Greater than 95% removal of iodine was achieved with sodium tetraborate in the ice.

- The physical form of the ice did not appear to have a strong influence on iodine removal.
- The effect of iodine concentration was small.
- Iodine removal was observed to be a strong function of air in the steam/air mixture.
- The ice condenser was not effective in the removal of methyl iodide.

No consideration was given to the retention of other gaseous and particulate fission products, nor were the effects of high concentration aerosols from core/concrete reaction considered.

The continued effectiveness of an ice condenser system in the removal of fission products following a severe accident depends on the circulating fan system remaining operational during exposure to the dense aerosols and other debris generated during the accident. No information could be found either confirming or denying the long-term operability of this fan system under the type of conditions following a severe accident.

2.2.3 Containment Sprays

The containment spray system (CSS) is a part of the residual heat removal (RHR) system and provides containment cooling following a loss of coolant accident in addition to being a fission product removal mechanism. The CSS is supplied with water from the suppression pool, containment sump, or refueling water storage tank, depending on reactor type and the conditions following the accident. In some plants, the spray fluid is a dilute sodium hydroxide or hydrazine solution which enhances the ability of the spray to remove iodine in the containment atmosphere. The ability of the spray system pumps to continue to function following the incident is addressed in Section 2.2.4.

Because of the large surface area between the spray droplets and the containment atmosphere, the containment spray serves as an effective means of removing fission products following an accident. The major benefit of the containment spray is its capacity to absorb molecular iodine and aerosols from the containment atmosphere and thereby reduce their release to the atmosphere. Although, the spray will also slowly remove organic iodine forms, this is not always considered in spray system performance analyses.

Experimental evidence on the performance of containment spray systems was provided by the CSE project at Hanford (Hilliard and Postma 1980, 1981). These tests considered the removal of a simulated fission product consisting of UO_2 , elemental iodine, methyl iodide and Cs. This simulant was introduced into the test vessel containing an air-steam mixture. In most tests, natural processes were allowed to attenuate the fission product inventory during the initial 1/2-hour period before the sprays were turned on. In two tests, the spray was started before the iodine was released. These experiments were designed to test models of elemental iodine washout, but the significant removal of cesium and UO_2 particles was also demonstrated. The initial application of spray provided the greatest reduction in iodine concentration and subsequent applications gave progressively slower washout rates as the concentration in the vessel was reduced. Typical spray attenuation factors of 0.033 were observed for the first 2-hour period, and 0.010 after 24 hours. It was concluded that sprays were more effective than filter systems for the removal of airborne iodine and greatly exceeded the performance of natural removal mechanisms during the first 2 hours following the release. Over a 24-hour period, the removal rate for the natural processes compared more favorably with the engineered safety features.

2.2.4 Pumping Systems

The residual heat removal (RHR) and containment spray (CS) systems are supplied by pumps which take suction from either the pressure suppression pool or the containment sump. There has been some concern raised regarding the ability of these pumps to continue to function following an incident because of the debris that will be generated by the incident and carried to these water supplies. This debris could possibly plug the inlet screens or cause accelerated wear in the pump seal. At Creare, Inc. the performance of the RHR and CS pumps was studied for a sample of power reactors under conditions of debris and air ingestion expected after a LOCA (NUREG/CR-2792). The reactors included:

- Arkansas 2
- Calvert Cliffs 1 and 2
- Crystal River 3
- Kewanee
- McGuire 1 and 2
- Midland 2
- Millstone 2
- Oconee 3
- Prairie Island 1 and 2
- Salem 1.

The analysis was based on experimental data on pump performance and net positive suction head and data in literature regarding the performance of pumps subjected to debris and air ingestion. Several types of debris were considered, and conservative estimates were made of the quantities of each that would pass through the pump:

• Insulation fibers (0.3% volume concentration)

- Aluminum and zinc hydroxide precipitates (0.04% volume concentration)
- Paint flakes (0.025% volume concentration)
- Concrete dust (0.05% volume concentration).

The total concentration of debris reaching the pumps was estimated to be less than 0.5%. The investigators concluded that the hydraulic performance degradation of RHR and CS pumps should be negligible and mechanical wear should be too small to seriously impair long-term pump operation. If failure of the shaft seals should occur, leakage would be less than 0.1% of pump flow rates.

Burns and Roe, Inc. (NUREG/CR-2791) investigated the possibility of debris blocking the pump inlet screens enough to degrade pump performance. Reactors specifically examined included:

- Salem 1
- Arkańsas 2
- Main Yankee
- Sequoyah 2
- Prairie Island 1.

The debris was assumed to be principally insulation generated by pipe whip, pipe impact and jet impingement. The amount of debris estimated was found to depend on the plant and type of break and ranged from 100 to 7200 ft³. The results of the analysis were not sufficiently conclusive to generalize whether screen blockages are sufficient to cause pump malfunctions. Depending on the reactor and type of break, screen blockage ranged from 0 to 100 percent. It was concluded that the question of screen blockage must be addressed on a reactor-by-reactor and accident-by-accident basis.

Neither study addressed the potential problems caused by the debris generated by core/concrete interactions following a severe accident.

2.2.5 Filter Systems

Filter systems are used as atmosphere cleanup systems in several ESF systems. Systems of particular concern in severe accident analysis include the secondary containment filter systems and the containment air recirculating systems.

The secondary filter systems are employed to clean up contaminated air that escapes from primary to secondary containment following a LOCA. These systems are given various names by the various reactor vendors including the "auxiliary building filter system, the "standby gas treatment filter system", the "emergency gas treatment system", "penetration room exhaust system" and the "secondary containment air cleanup system."

In these systems, air is collected by ducts from regions where primary leak potential exists and it is moved by fan through a filter train and up through the containment building exhaust stack. The filter system removes fission products from the air before the air is released to the environment. In some plants, there is an annulus between the containment building and the shield building where the atmospheric air is passed through a similar filter system and at least partially exhausted to the environment.

Typically, the filter system consists of a train of equipment, including a moisture separator, heater, prefilter, high efficiency particulate air (HEPA) filter, charcoal adsorber trap, and another HEPA filter. After passing through this train, the filtered air is exhausted by the fan through the containment building stack. Air flow rates through a filter train are typically in the 1000- to 10,000-cfm range at 5 to 10 in. wg. There are two parallel filter trains so that backup capability exists.

The moisture separator (or demister) removes entrained water droplets and moisture from the air stream. The removal water flows into a drain at the bottom of the separator. The electric heater unit heats the air to reduce the relative humidity to 70% or less, because higher humidity air would reduce the effectiveness of the downstream HEPA and charcoal adsorber filters.

The following prefilter removes the larger particles and with about 85% efficiency. The smaller particles that get through are collected on the adjacent HEPA filter. The HEPA filter collects up to 99.97% of the particulates down into the 0.3- μ m-dia size range. The HEPA filter is water and fire resistant and is capable of functioning to 500°F with gamma doses to 10⁶ rads.

Gases that exist the HEPA filters are trapped in the charcoal adsorber unit. The adsorber is made up of charcoal impregnated with potassium iodide. This adsorber removes up to 99.9% of the methyl iodine and elemental iodine in 70% relative humidity air at 175° F. These is concern about the charcoal exceeding its ignition temperature of 644° F because of decay heat generated by the highly radioactive elements collected in the charcoal. A temperature sensor and cooling spray is installed in the unit to cool the charcoal and prevent fires from starting.

The downstream HEPA filters collect particulates which might be released from the charcoal adsorber unit. The fan transfers the filtered air to the containment building stack where it is released to the environment. A standby cooling fan is available in case the main fan is shut down or otherwise inoperable.

Containment Air Recirculating Systems are used in some of the earlier PWRs to remove fission products within the containment following an accident. These systems include moisture separators, prefilters, HEPA filters and activated charcoal adsorbers. Following an accident, the air flow is divided with 30% passing through the filter and the remainder through a cooling system. The vapor flowing through the air coolers will tend to condense and the resulting liquid water will tend to trap some fission products. However, no experimental information quantifying this removal mechanism could be found.

The filter systems are intended to trap iodine (elemental and organic) and aerosols. However, they are not designed to accommodate large quantities of aerosol materials. One HEPA filter (0.47 m^3/s) will trap only 1 kg of aerosol material before being effectively plugged (NUREG-0772). Therefore, this system could become ineffective under accident sequences involving core melting where large amounts of aerosols (above 500 kg) would be generated (NUREG-0772).

The design, construction, and testing of ESF filter systems are highly formalized and subject to the requirements and recommendations of NRC Guides, codes, and standards (Regulatory Guide 1.52; ANSI N510-1975; ANSI N101.0-1972; Hilliard and Postma 1981). The ESF filter system designs are subject to the regulations of NRC Regulatory Guide 1.52. HEPA filters are shop tested according to MIL-STD-282. Impregnated activated carbon is tested prior to installation for elemental iodine and methyl iodide removal, apparent density, hardness, moisture, particle size distribution and ash content, surface area, impregnate content, leachout and ignition temperature. It appears that the ESF filter systems are very effective in removing elemental iodine and organic iodide, and the industry appears well equipped to test and verify the performance of these systems.

A series of tests in the CSE facility at Hanford demonstrated the effectiveness of ESF filters (McCormack et al.). Five tests were performed with an internal recirculating filter-adsorber loop consisting of a prefilter demister, a HEPA filter and charcoal bed in series. The units were nominally designed for 1000 ft³/min. It was found that the filter systems were very effective for the removal of iodine compared with water sprays and natural removal mechanisms, especially during the first 1/2 hour of the test, because of their superior capability to remove methyl iodide. Even after 24 hours in a steam-air environment, the filter units were observed to remove methyl iodide at nearly the initial rate observed for a new unit. Typical attenuation factors for the filter loop were 0.13 for the initial 2-hr periods and 0.013 for a 24-hr period. Although these CSE test show that filters are very effective in the removal of aerosol in the containment, the aerosol concentrations used in these tests were small in comparison to many severe accident sequences. If more realistic concentrations were used, the filters would have swiftly plugged.

2.3 ASSESSMENT OF THE ESF SYSTEMS DATA BASE

The present data base on the performance of ESF systems in removing airborne fission products following a severe accident is adequate in some areas and very sparse in others.

There appears to be a large body of data available on fission product retention by subcooled and pressure suppression pools. However, most of these data were obtained under conditions not representative of degraded core accidents from the standpoint of release rates, pool depths or particle size distributions. Also, no information is available on CsI scrubbing. However, General Electric Co. has recently completed pool scrubbing tests and has updated the results of these tests in proprietary documents. Also, BCL recently undertook an experimental program for EPRI on the removal of radionuclide aerosols in BWR suppression pools and PWR quench tanks. It is hoped that these tests will provide the data required to support a realistic scrubbing model for reactor safety analyses. Large-scale tests currently being planned for the Marviken reactor may provide some of the data required for the verification of new and existing models.

The function of the Westinghouse ice condenser system in attenuating the pressure rise in the containment vessel following a LOCA is well documented. However, data on the removal of fission products is not extensive. Some information is available on the influence of the following variables on elemental iodine removal from ice condenser influent: 1) steam concentration, 2) ice additives, 3) ice loading, 4) vapor temperature, 5) flow velocities and 6) iodine concentration. Other information is also available to indicate the ineffectiveness of the ice condenser in removing organic iodine. However, little is known about the ice condenser's effectiveness in removing other soluble and insoluble fission product aerosols. Neither is much known about the continued effectiveness of the ice condenser when heavily loaded with aerosol materials such as those that could occur following vessel melt-through and concrete/core interaction. No information is available on the continued effectiveness of the ice condenser air circulation system in the presence of the debris generated by a severe accident.

The ability of sprays to remove elemental iodine can be modeled by standard absorption theory, and the models have been confirmed by large-scale experiments. Aerosol scrubbing by sprays is also reasonably well understood, and verified models are available to describe capture by the several mechanisms involved. However, the available data do not cover the ranges of steam flux, forced- and natural-convection velocities, and particulate aerosol concentrations expected during accident conditions. Also, the influence of high droplet loadings of soluble and insoluble materials on the scrubbing performance of sprays is unknown.

It appears that there is reasonable confidence that the debris generated during loss-of-coolant accidents would not cause a significant degradation of the RHR and CS pumps through mechanical wear or shaft seal failure. However, the possibility that debris generated by a severe accident would block the pump inlet screens sufficiently to degrade pump performance has been shown for several reactors under certain LOCA conditions. No information is available on the effect of the type and amount of such debris on the continued satisfactory operation of these pumps. Based on the earlier studies of LOCAs, the influence of this debris is reactor specific and must be considered on a reactor-byreactor basis.

Filter systems are designed on the basis of rather formal NRC Guidelines for specific design basis accidents. They are arranged so that they can be tested in place with DOP (dioctylphtalate) or DEHS aerosol and Freon. A great deal of work has been done to characterize the performance of air cleaning devices, and the available information is adequate to predict performance under accident conditions. Little information is available regarding the failure modes of HEPA filters or charcoal adsorbers. Such failures may occur as a result of hydrogen combustion, heat generated by entrapped fission products, or rupture caused by increased pressure drop. Failure of these devices may re-release large quantities of entrapped aerosol materials to the containment.

The air coolers in Containment Air Recirculating Systems have the potential for removing airborne fission products because they will be wetted by condensing steam from the containment atmosphere. No information is available to quantify this removal means, and no work on this topic is currently under way.

Fans driven by electric motors are used in containment air coolers, filter systems and ice condenser systems. These systems require the continued operation of these fans following a severe accident for the effective removal of fission products. However, it is possible that high containment temperatures, high-density aerosols, and debris resulting from these accidents may interfere with the operation of these fans or cause premature failure. No information was found that would indicate whether this would be a problem or not.

3.0 ADDITIONAL DATA BASE REQUIREMENTS

The purpose of this section is to define experimental data requirements to support the development and verification of mechanistic models that describe fission product removal by ESF systems. As part of the discussion, some of the existing and developmental models are briefly reviewed for each of the ESF system types: 1) Suppression pools, 2) Ice beds, 3) Containment sprays, 4) Filter Systems, 5) Containment Coolers.

3.1 SUPPRESSION POOLS

3.1.1 <u>Models for Predicting the Fission Product Removal Performance</u> of Suppression Pools

Most published models for analyzing pool scrubbing are non-mechanistic and express fission product scrubbing parametrically in terms of a "decontamination factor" (DF).

Both Diffey et al. (1965b) and Devell et al. proposed models for the scrubbing of elemental iodine. Diffey assumed that the iodine in the bubbles leaving the water pool was in equilibrium with the iodine in water. Devell's model improved upon Diffey's by considering the degree of equilibrium that would be achieved between the bubbles and the water. The Diffey model expressed the absorption of elemental iodine in terms of the volume of gas bubbles leaving the pool, the volume of the water pool, and the iodine partition coefficient. Additional variables considered by the Devell model included submergence depth, bubble rise velocity, bubble volume, bubble surface area, and mass transfer coefficient.

Postma et al. (NUREG/CR-2659) and Raghuram et al. developed models for the scrubbing of iodine in bubbles in steam generators. The models conservatively considered only the effect of gravity settling and expressed the scrubbing in terms of 1) particle settling velocity, 2) bubble diameter, and 3) bubble rise time.

Fuchs described a model for particle scrubbing that accounted for gravity settling, inertial deposition due to centrifugal force and diffusional deposition. Variables influencing the scrubbing included 1) particulate size distribution, 2) bubble diameter, 3) bubble rise velocity, 4) bubble rise time, 5) temperature, and 6) pressure. NUREG-0772 recommends two simple models for the scrubbing of iodine vapors and particles. These are similar to the models proposed by Postma et al. (NUREG/CR-2659) and Raghuram et al.

None of the above described models are completely satisfactory for assessing the performance of suppression pools because potentially important phenomena have not been taken into account. These phenomena include: 1) slow chemical reactions that may enhance the retention of volatile materials such as I_2 , 2) bubble circulation effects, 3) particle growth, 4) diffusiophoresis 5) thermophoresis, 6) diffusion, and 7) entrance effects.

New theoretical models for pool scrubbing were recently developed by Science Applications, Incorporated (SAI) and the Pacific Northwest Laboratory. Publication of these models is expected in the near future.

3.1.2 Data Requirements for Pressure Suppression Pools

A suitable data base to support the development and verification of a mechanistic model to describe the removal of aerosols by a suppression pool should include consideration of the following parameters over the range of values expected during the various hypothesized accident sequences:

- chemical composition of volatile fission products
- equilibrium partition coefficients for volatile fission products
- particle chemical composition
- particle density and aerodynamic size distribution
- steam fraction in the inlet gas
- gas flow rate
- temperature and pressure of inlet gas
- pool water temperature
- volume of pool
- downcomer submergence depth.

In addition, some consideration should be given to the effects of the loading of the pool water with aerosol materials. The BCL/EPRI large-scale experimental study is planned to address the influences of many of these parameters and may resolve some of the outstanding issues. However, more information is required on the details of the test plan to determine if this program really addresses all of the crucial issues and if it will provide confirmatory data for the model currently under development. Assuming it does or can be modified to do so, there appears to be little need to initiate additional experimental activities in this area in the near future.

3.2 ICE BEDS

3.2.1 Models for Predicting the Fission Product Removal Performance of Ice Beds

No mechanistic models of fission product scrubbing are known to exist. However, such a model is under development at PNL and the initial results are expected to be published in 1983. During this work, the following mechanisms were identified as important in the removal of aerosol particles:

- gravity settling on structural and ice surfaces
- impaction and interception by the ice and structural components
- diffusional deposition
- turbulent deposition on ice surfaces
- o diffusiophoresis
- thermophoresis
- particle growth from water vapor absorption.

The following two mechanisms appear to be important for the removal of elemental iodine:

- absorption by water from melting ice
- deposition on structural surfaces.

3.2.2 Data Requirements for Ice Beds

Since the data base for the removal of iodine by ice beds is limited and the data base for the removal of particulates virtually non-existent, it is important that an experimental program be undertaken to develop suitable information to support the model currently under development. Such an experimental program should include considerations of the influences of the following parameters on fission product removal:

- chemical composition of particles
- chemical composition of volatile fission products
- particle size and aerodynamic size distribution
- steam fraction in gas
- temperature and pressure of inlet gas
- quantity and configuration of ice available

- gas flow rate through ice compartment
- chemical composition of ice.

The experimental apparatus should be on as large a scale as possible to eliminate questions of scale and should preferably use prototypical hardware.

3.3 CONTAINMENT SPRAYS

3.3.1 Models for Predicting the Performance of Containment Sprays

Aerosol scrubbing by sprays is reasonably well understood and several models are available to describe the several processes involved.

The SPIRT code (Postma et al. 1978) is used by NRC to conservatively predict the washout of elemental iodine and methyl iodide in source term calculations. Spray processes considered by SPIRT include:

- droplet coalescence
- iodine washout
- methyl iodide washout by thiosulfate sprays through absorption and chemical reactions.

Particle washout is not included in published versions of SPIRT. Key variables influencing the scrubbing process are 1) iodine partition coefficient, 2) drop fall distance, 3) drop size distribution, 4) spray flow rate, 5) atmosphere temperature and pressure, 6) spray temperature, and 7) reaction rate constant for methyl iodide. The CORRAL code was written to yield realistic predictions of fission product washout for reactor safety studies (Battelle Columbus Laboratories). It uses a "well-mixed" drop model to analyze the washout of elemental iodine and an empirical model for aerosol particles based on the CSE tests. Key parameters in the CORRAL code include 1) drop fall height, 2) mean drop diameter, 3) spray flow rate, 4) atmosphere temperature and pressure, 5) volume of gas phase, and 6) spray composition.

A number of accident analysis codes are currently under development or are being modified to include spray washout models. These include MATADOR (BCL), CONTAIN (Sandia), TRAP-MELT (BCL) and NAUA-4 (KfK).

3.3.2 Data Requirements for Containment Sprays

The most important parameters that should be characterized in the containment spray data base include:

- aerosol particle size and density
- spray drop size, flow rate and fall height
- atmospheric temperature and pressure

- gas flow rate
- steam content of gas.

There is also some question regarding the effect of recirculated spray water already heavily loaded with aerosol materials on the removal effectiveness of the spray.

Although a considerable quantity of data is available from the CSE experiment on aerosol removal by containment sprays, it does not cover the total range of variables expected in postulated degraded core accidents. Therefore, expansion of the current data base is required.

One matter that should probably be pursued further is the possibility of CS pump failure caused by inlet screen plugging. Existing studies should be extended to consider aerosol concentrations more representative of severe accidents to assess the potential seriousness of this problem.

3.4 FILTER SYSTEMS

3.4.1 Filter System Analysis Methods

Filter systems (particulate filters and charcoal adsorbers) are designed and analyzed using rather formal and detailed NRC guidelines. Their performance is rather well defined, and manufacturers' test data as well as in-situ performance data are available. The technology appears to be available to adequately predict the fission product capture efficiency under the various accident sequences.

3.4.2 Data Requirements for Filter System Performance

No immediate need for further performance data is seen because:

- The performance of such system is already well defined.
- Recirculating filter systems that could affect containment system contamination are found principally on the older plants.
- These systems would quickly plug during many serious accident sequences, which would render the systems ineffective.

There are some questions regarding the failure modes of prefilters, HEPA filters, and charcoal adsorbers as a result of such events as hydrogen combustion, the heating of entrapped fission products, or rupture caused by increased pressure drop. These events could cause the integrity of these units to be compromised, causing reentrainment of the aerosol materials already captured by the filter or adsorber. Data in this area are required.

3.5 CONTAINMENT COOLERS

3.5.1 Models for Performance of Containment Coolers

All reactors use ventilation equipment to control the containment air temperature during normal plant operations. Following a severe accident, this equipment may continue to operate, condensing steam in the containment atmosphere and removing entrained fission products. Although this function of containment coolers was observed in the 1965 incident in the Plutonium Recycle Test Reactor and the TMI-2 accident, no studies have been made on the effectiveness of this removal mechanism, and no models to describe the removal process have been developed.

3.5.2 Data Requirements for Containment Cooler Performance

Since no data apparently exists on the removal of fission products by containment coolers, an experimental program should be undertaken to provide a suitable data base for development of predictive models. Such an experimental program should consider the influence of the following parameters on fission product removal:

- chemical composition of volatile fission products
- chemical composition of particulates
- particle size
- particle concentration
- steam content in containment gas
- temperature and pressure of containment gas
- cooling coil surface temperature
- gas velocity through the coil
- coil dimensions (fin height, fin spacing, coil depth, etc).

Data should be obtained on the effects of aerosol material loading on the coal surfaces on the capture efficiency. Data should also be obtained on the total loading to effectively plug the coil.

This information would be of considerable interest since 1) all containment systems have these coolers, 2) the coolers operate all the time and require no special startup for accidents (operating conditions, however, may be changed following a LOCA), and 3) their performance in removing fission products has not been considered in past studies.

4.0 RECOMMENDED RESEARCH PROGRAM

This section describes recommended experimental activities to develop a data base for supporting the development and verification of models of ESF system fission product removal performance. Major experimental activities include:

- fission product scrubbing in ice condenser pressure suppression systems
- scrubbing of fission product aerosols by water sprays
- aerosol removal of air coolers
- interaction of aerosols with demisters
- influence of high-density aerosols on fan/motor performance.

In addition, several engineering studies and surveillance activities are recommended to help define and guide future analytic and experimental activities regarding the interactions of aerosols with ESF systems.

One of the most frequently recurring questions observed in the development of ESF/aerosol models and the application of these models to analyze severe accident situations regards the time-dependent composition (chemical and mechanical) of the aerosol interacting with the ESF systems for various accident sequences. Fission product release and transport studies are currently under way in both the United States and foreign countries and are aimed, in part, at better characterizing these aerosols. The consideration of these studies is not specifically within the scope of this present work. However, because of their importance to the development and application of ESF/aerosol models, the general recommendation is made that these studies be aggressively pursued to provide an improved data base and predictive capability for fission products and aerosols following a severe accident.

4.1 SUPPRESSION POOL STUDIES

The work currently under way at BCL sponsored by EPRI is expected to address many of the outstanding issues regarding the scrubbing of fission products by suppression pools. Therefore, it is not recommended that additional experimental work be undertaken by NRC in this area at the present time. However, it is recommended that NRC establish a task force with cooperation and participation of EPRI to follow the planning and progress of this work. The objective of this task force would be to maintain surveillance of all program activities to assure that all outstanding issues are adequately addressed and that the results will be of maximum benefit to the development and verification of computational models and codes. It is visualized that this task force would develop recommendations to NRC on additional activities that should be undertaken to obtain an adequate understanding of suppression pool scrubbing.

4.2 FISSION PRODUCT SCRUBBING IN ICE CONDENSER PRESSURE SUPPRESSION SYSTEMS

It is recommended that an experimental study be undertaken to develop a data base on the fission product aerosol removal performance of ice condenser systems under conditions representative of those following a severe accident. Data of particular interest include:

- flow patterns in and around the ice baskets
- available surface area and porosity of new ice, "aged" ice and ice that has been partially melted following a severe accident
- particulate agglomeration behavior following a severe accident
- aerosol removal behavior for both soluble and insoluble aerosols as influenced by particle size, particulate concentration carrier gas dynamics and ice conditions.

These data would be used to implement, improve, and verify predictive models of fission product scrubbing in ice condenser systems and to form a basis for the future development of advanced predictive methods.

It is recommended that this experimental program consider two principal test assembly types. The first should model one full-scale ice basket and associated adjacent gas flow passages. The second should model a full-scale section or sector of a typical ice condenser system. The first test assembly type would be used to obtain phenomenological data on the flow processes in and around the ice baskets, define the ice surface area available for the removal of particulates, quantify the contributions of the various removal mechanisms, and determine paticulate agglomeration behavior. Because this test assembly would contain only one ice basket, it would be possible to insert instrumentation at multiple locations along the ice basket and along the gas passages to study the detailed aerosol dynamics and adsorption mechanisms. This information would be useful in improvement of existing ice condenser models and to develop more realistic future models. The multiple basket section or sector model would be used to obtain data for the verification of these models.

The fission product aerosol simulants used in this program should be carefully selected and should include both soluble and insoluble particulates ranging in size from 0.5 to 10 μm .

It is recommended that the availability of the Westinghouse Waltz Mill Ice Condenser Test Facility be investigated for the multiple basket tests. If available, this facility could be modified and recommissioned for these experiments with a substantial savings of time and money over that required to build an entirely new facility.

4.3 SCRUBBING OF FISSION PRODUCTS BY WATER SPRAYS

It is recommended that the CSE water scrubbing tests performed in the late 1960s be extended to consider a broader range of variables for the purpose of

validating and improving existing spray removal models. Specific variables that should receive expanded attention are

- aerosol concentrations
- aerosol chemical composition and particle sizes
- water spray operation sequence
- steam/fission product release schedule.

Testing should make use of improved aerosol simulants developed in more recent fission product transport experiments.

The CSE facility should be examined as a potential location for these recommended experiments.

4.3.1 Containment Spray Inlet Screen Plugging

It is recommended that an engineering study be undertaken to determine the effect of accident debris on the degradation of CS pump performance by inlet screen plugging. In earlier studies, it was found that some reactors could experience problems with the quantity of debris generated by a LOCA. Since the loss of these pumps could have a serious effect on the source terms, these studies should be extended to consider the types and quantities of debris resulting from a severe accident. Because of variations in the design features between reactors, this study should consider all reactors in service and under construction.

4.4 FILTER/COOLING SYSTEMS

Air coolers and filter systems consisting of moisture separators, prefilters, HEPA filters and charcoal adsorbers are used in a variety of system configurations and can vary from reactor-to-reactor. Although it was possible to identify experimental studies to address several generic issues dealing with these systems, it was not possible to adequately identify all potential issues because of the large number of existing system designs and the limited information on how aerosols interact with elements of these systems under severe accident conditions. It was particularly difficult to identify interactions between system components as they become effectively plugged with aerosol materials. Also, it was difficult to identify failure mechanisms of system components, especially HEPA filters and charcoal adsorbers, from mechanical and thermal loading of entrapped materials and from the combustion of hydrogen. Therefore, it is recommended that an engineering study be undertaken to identify typical systems and to assess the interaction of these systems with aerosols under conditions expected during a severe accident. Special emphasis should be placed on examining potential failure modes of filters and charcoal adsorbers including the effects of hydrogen combustion, fission product heating, and mechanical loading from wet, entrapped aerosol materials. The results of this engineering study would be used to better define the outstanding

questions regarding ways these systems behave in the presence of dense aerosols and to provide a basis for developing additional experimental work to address these questions.

Sections 4.4.1, 4.4.2, and 4.4.3 describe several generic-type studies to address specific questions identified during the present study.

4.4.1 Aerosol Removal by Containment Air Coolers

The purpose of this suggested program is to develop data on the fission product removal by reactor containment air coolers. This data will be used to support development and verification of models to predict the behavior of these units following postulated severe accident involving core melting. Specific information that would be obtained include:

- rate of removal of soluble and insoluble aerosols as influenced by 1) the heat transfer parameters, 2) aerosol composition (chemical and particle size), and 3) loading
- rate of removal of gaseous fission product simulants
- total removal capability (loading) before units become effectively plugged.

This work should include a survey of existing literature on the fouling of extended-surface heat exchangers to determine the applicability of a large body of available information on the deposition of particulates (fly ash) in fintube economizers and air heaters. The experimental activities should emphasize the types of air cooling coil configurations currently in use in containment air cooling systems. Sufficient additional configurations should be considered to enable definition of the effects of fin height, fin spacing, tube spacing and coil depth on capture performance. It is envisioned that 15 to 25 coil configurations may be required to adequately cover the range of configuration details and dimensions. However, the test assemblies can be small, typically with 1 to 4 ft² of face area and full depth in the direction of air flow. The results from these small test assemblies can be confidently scaled up to full size for application of the data to accident analyses.

Steam will be injected into the air flow upstream of the test assembly to simulate post-accident atmospheres in the containment. Simulants will be introduced to represent gaseous fission products, soluble aerosols and insoluble aerosols. The method of producing these simulants will be consistent with the best current practice in on-going fission product transport studies.

Experimental conditions will be chosen to statistically cover the range of thermal hydraulic parameters anticipated during postulated degraded core accidents. The fission product simulants will be introduced individually and in combination to investigate the influence of 1) chemical composition, 2) particle size, 3) particle density, and 4) surface loading on the capture rate. Samples of the air/steam/simulant mixture will be taken before and after the test assembly by filter or impactor samplers. Measurements will be made of the

thermal hydraulics conditions upstream and downstream of the test assembly, pressure loss, and heat load on the test coil. Condensate formed will be collected and analyzed for simulant content. Periodic examinations of the coil surfaces will be made to characterize simulant deposition, and the time to effective plugging of the test assembly will be determined.

4.4.2 Influence of High-Density Aerosols on Fan/Motor Performance

Electric motor-driven fans are associated with containment air coolers, filter systems and ice condenser systems. Fan operation imposes acceleration fields on aerosol-laden air passing through them, which may cause dropout of aerosol materials. Accumulations of these materials may cause obstruction of flow paths and, possibly, overloading of the drive motors caused by interference with moving parts. The totally enclosed, externally cooled drive motors used in many of these systems also contain an integral centrifugal fan, which is subject to similar plugging if exposed to a high-density aerosol. Such plugging could impede cooling of the motor, and coupled with high containment temperatures following a severe accident, it could lead to subsequent overheating and failure. It is recommended that experiments be performed to expose typical fans and fan motors used in these ESF systems to high-density aerosols to determine the influence of these aerosols on performance, and particularly on the post-accident operational lifetime of these units. Such a study could be integrated into the aerosol transport studies presently under way at ORNL or Sandia.

4.4.3 Aerosol Removal by Demisters

Filter systems nominally contain demisters as one of the initial elements in the equipment train. Since these elements are designed to effectively eliminate a water aerosol from the flowing air, they will also effectively remove the high-density aerosols resulting from vessel failure and core/concrete interactions. Although the total removal capabilities of demisters is not judged to be significant, the period of time it takes to plug them may be important since such plugging will render the remaining filter components ineffective for the removal of aerosol materials. It is suggested that the removal behavior of demisters be studied. This study should consider the principal demister configurations in common use. The data obtained should be sufficient to support the development of a removal model and predictions of the plugging behavior. It is recommended that this task be integrated in the study described in Section 4.4.1. •

5.0 PEER REVIEW

Following the preparation of Sections 1 through 4 of this report, the report was submitted for peer review by a group of researchers working in the areas of 1) the source term definition and 2) fission product removal performance of ESF systems. These researchers are listed in Section 5.1. They were requested to review this document and provide comments with particular emphasis on the adequacy of the proposed experimental program defined in Section 4.0. Also, they were requested to suggest additional experimental activities that they felt necessary to address specific problem areas associated with fission product removal by ESF systems. The purpose of this section is to describe the results of this peer review.

5.1 THE PEER REVIEW GROUP

Members of the peer review group and their affiliations were as follows:

R. S. Denning (Battelle-Columbus)

J. A. Gieseke (Battelle-Columbus)

R. K. Hilliard (Hanford Engineering Development Laboratory)

T. S. Kress (Oak Ridge National Laboratory)

D. A. Powers (Sandia National Laboratories)

R. L. Ritzman (Science Applications, Inc.)

R. C. Vogel (Electric Power Research Institute

F. J. Rahn (Electric Power Research Institute)

Comments regarding this document were received from the peer group members by letter and telephone. Copies of these letters and telephone contact memoranda will be retained by the authors for future reference or documentation.

5.2 RESULTS OF THE PEER REVIEW

All comments received from the peer review group were very positive in all respects. Comments on the state-of-the-art review sections of the report included the following:

"...it is a valuable document with a good review of the state-of-the-art in this area" (EPRI) "The report is basically well organized, well written, and a good treatment of the Technical topic under study." (SAI) "...good document - right perspective." (EPRI)

Several reviewers provided editorial comments, technical corrections and additional information to augment that presented in the first draft of this

report. All of these comments, corrections and information were incorporated in the final draft. One comment was made that the review of fission product releases and chemical forms of these releases was "...very brief and incomplete." Because this section was originally intended only as a brief overview of this subject for the purpose of introduction, no further revision of this section was performed.

The research program described in Section 4.0 was generally well supported by the reviewers responding. No additional experimental or engineering studies were recommended. Comments provided include the following:

"...we feel it proposes a balanced research program" (EPRI). "I have no tests or studies to recommend beyond those covered in the report." (SAI)

R. L. Ritzman (SAI) provided two additional comments:

- "I question the cost effectiveness of the large-scale (ice condenser) verification tests. Unless the Westinghouse facility could be used in a straightforward manner, the tests are excessively expensive."
- "I also have some reservations about the need for the further spray testing suggested in this section. It should not be considered a high-priority item."

The authors have also recognized the potential high costs of the proposed ice condenser verification tests. Efforts are under way at PNL to determine the availability of the Westinghouse facility for this work and to identify alternatives if this facility is not available. The comment regarding the priority of the proposed spray tests supports the observations made in Section 2.0 of this report that the data base already available to verify spray models is quite extensive. However, data in the range of variables important to severe accident analysis are somewhat sparse. The authors believe that it is important that adequate data be obtained to validate spray codes under conditions that the codes will be used. However, there is the basic agreement that the other proposed tasks have sufficiently greater importance that they should be given a somewhat higher priority.

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reactor Engineered Safety Feature (ESF) system systems considered included pressure suppressi containment sprays, filter systems and contain assessment, a research program was recommended the development of mechanistic models and compo- system fission product removal. This research to better define the performance of ice conden- available on water spray systems and to invest coolers, demisters and fans in the presence of following a severe accident.	removal s was rev on pools, ment air o to expand uter codes program ser system igate the aerosols	capabilities iewed and ass ice condense coolers. Bas d this data b s for the pre included expe ns, expand th behavior of typical of t	of nuclear sessed. The er systems, sed on this base to support ediction of ESF erimental efforts he range of data containment air chose expected		
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