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Licensing Design Basis Source Term Update for the Evolutionary Advanced Light Water Reactor

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Department of Energy

***Advanced
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**LICENSING DESIGN BASIS SOURCE TERM UPDATE FOR
THE EVOLUTIONARY ADVANCED LIGHT WATER REACTOR**

Prepared by
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ABSTRACT

The purpose of this report is to document the technical basis for a licensing source term update for the Evolutionary Advanced Light Water Reactor (ALWR) which will make the source term more physically realistic. While TID 14844 and related regulatory guidance have served the industry well, much has been learned about source term over the last 30 years, and the ALWR Requirements Document provides an opportunity to incorporate this experience by updating the licensing source term. Further, the source term update will provide an improved basis for Evolutionary ALWR accident mitigation design.

Results of this work indicate that the fission product release magnitude to containment is slightly less than TID 14844 for noble gas, iodine, and semi and low volatiles, but somewhat higher for cesium and tellurium. Release timing is delayed by one hour or more after the accident initiation. The chemical form of iodine is largely aerosol with significantly less organic iodine compared to regulatory guidance which specifies mostly elemental and a relatively large fraction of organic. Containment spray aerosol removal rate was determined to be significantly higher than specified in regulatory guidance. Finally, BWR suppression pool decontamination factor was determined to be less effective than allowed by regulatory guidance early in the accident (due to the delayed release noted above) and more effective than that allowed by regulatory guidance later in the accident.

It is recognized by the ALWR program that the source term update could be taken further in the direction of a physically-based source term. Schedule and resource constraints have prevented doing this for the evolutionary plant, although such an effort is underway for the passive plant. Notwithstanding the schedule and resource constraints, the work reported here, while in the nature of a progress report, is considered to be a useful update of existing source term regulatory guidance.

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ACRONYMS AND ABBREVIATIONS

ALWR	Advanced Light Water Reactor
ARSAP	Advanced Reactor Severe Accident Program
BWR	Boiling Water Reactor
CFR	Code of Federal Regulations
DBA	Design Basis Accident
DOE	Department of Energy
ECCS	Emergency Core Cooling System
EPRI	Electric Power Research Institute
Exp. Cond.	Experimental Condition
IDCOR	Industry Degraded Core Rulemaking
INEL	Idaho National Engineering Laboratory
LACE	LWR Aerosol Containment Experiment
LOCA	Loss of Coolant Accident
LOFT	Loss of Flow Test
LPIS	Low Pressure Injection System
LWR	Light Water Reactor
MAAP	Modular Accident Analysis Program
NRC	Nuclear Regulatory Commission
NYPA	New York Power Authority
ORNL	Oak Ridge National Laboratory
PBF	Power Burst Facility
PRA	Probabilistic Risk Assessment
PWR	Pressurized Water Reactor
RCS	Reactor Coolant System
SFD	Severe Fuel Damage
SRP	Standard Review Plan
STEP	Source Term Experimental Program
TID	Technical Information Document
TMI	Three Mile Island

1.0 INTRODUCTION

1.1 Background

The purpose of this report is to document the technical basis for a licensing source term update for the Evolutionary Advanced Light Water Reactor (ALWR) which will make the source term more physically realistic. The report has been prepared by the Department of Energy (DOE) sponsored Advanced Reactor Severe Accident Program (ARSAP) in support of the Utility/Electric Power Research Institute (EPRI) ALWR Program.

Since the early 1960s the nuclear industry has used TID 14844 as the basis for fission product release in the source term used for siting dose evaluations and other applications. While TID 14844 and related regulatory guidance has served the industry well, resulting in a strong containment and engineered systems for accident mitigation, much has been learned in the last ten years from analysis of the Three Mile Island Unit 2 (TMI-2) accident and subsequent severe accident research. The ALWR Requirements Document provides an opportunity to incorporate this experience by updating the licensing source term.

A review of 10CFR100 indicates that TID 14844 is "guidance in developing the exclusion area" and "may be used as a point of departure" for considering site requirements.¹ Regarding the question of whether an amendment to 10CFR100 (i.e., a rulemaking) is necessary to update the licensing source term, NRC's General Counsel stated in a Commission meeting that "the regulation itself would not have to be changed" to use the more current source term information.² Thus it is assumed in this report that reasonable, technically justified modifications can be made to the source term through changes to regulatory guidance as opposed to amending 10CFR100. The NRC staff indicated in a presentation to the Advisory Committee on Reactor Safeguards on February 9, 1990 that evolutionary plants are to meet 10CFR100 and that engineering judgment will be used to allow deviations from classical source terms in safety evaluation reports.

There are two main objectives in updating the licensing source term. The first is to factor in the source term experience of the last decade as

noted above. The second objective is to provide a more rational basis for Evolutionary ALWR accident mitigation design. Progress on the Evolutionary ALWR source term update work is being reported here as far as it has been taken. Resource and schedule constraints have prevented taking this work further in the context of the evolutionary plant. A more complete source term update is now being pursued by the ALWR Program for the passive plant.

1.2 Licensing Source Term Applications

An important factor in the consideration of change to the licensing source term is the application to which the source term is put. It is certainly a bounding assumption for airborne releases, for example, to assume that all fission products are released from the reactor coolant system (RCS) into the vapor space of the containment, and none into the post-accident liquid phase. This assumption, however, is not useful in the assessment of the radiation doses of equipment exposed to the liquid phase in the post-accident containment environment. Similarly, the assumption of an instantaneous release of a large fraction of the core inventory may be useful for the calculation of two-hour off-site doses, given that the dose "clock" also is started "instantaneously", i.e., at the time of the initiating event. This assumption, however, is not appropriate when applied to mitigative system design, such as the spray additive system, where it can result in unnecessary system complexity, or a design that performs at less than an optimum level under realistic conditions.

In this discussion of licensing source terms, it is recognized that the TID 14844 fission product release, although originally intended for siting dose calculations, has found application as the design basis for a number of systems, such as containment spray system, spray additive system, stand-by gas treatment system, filtration systems, leakage collection systems, and control room habitability systems, as well as serving as an acceptability criterion for post-accident instrumentation and equipment qualification. A list of applications and the corresponding regulatory guidance documents are shown in Table 1.

TABLE 1. RELATIONSHIPS BETWEEN PLANT FEATURES AND REGULATORY GUIDANCE
BY THE DBA-LOCA SOURCE TERM

Topic	Regulatory Guide	Standard Review Plan Section
Offsite radiological consequences	1.3, 1.4, 1.7	15.6.5 A, B, D
Containment sprays	1.3, 1.4, 1.7	6.5.2, 15.6.5.A
Containment recirculation filters	1.3, 1.4, 1.52	6.5.1, 15.6.5A
Auxiliary building filters	1.52	6.5.1, 9.4.2, 3, 4
Main Steam Isolation Valve Leakage control	1.3, 1.96	6.7, 15.6.5.D
Standby gas treatment	1.52	9.4.5, 15.6.5
Ice condenser	-	6.5.2, 3, 4
Containment leakage	1.3, 1.4	6.2.1, 6.2.6, 6.5.3
Dual containment	-	6.5.3
Pressure suppression pool	-	6.5.3
Control room Habitability systems	-	6.4
Postaccident environment	1.89, 1.97	6.1.1, 6.1.2, 9.3.2
Emergency planning	-	13.3

It is the intent of this report to focus on the source term from a core melt accident as applied to offsite dose calculation as required in 10CFR100 and the systems which mitigate this offsite dose. It is recognized that, prior to implementation of any changes to the source term, a thorough review of licensing source term applications in addition to offsite dose will have to be made to ensure that the updated source term is appropriate for the intended application.

1.3. Areas of Licensing Source Term Update

The areas in which licensing source term changes have been developed for the evolutionary plant are as follows: fission product release timing, release magnitude to the RCS, fission product chemical form, RCS retention, and fission product removal in containment. Evolutionary ALWR severe accident sequences have not been analyzed in this work, but rather it was assumed for the licensing source term update, as far as it was taken, that "substantial meltdown" took place without defining how it occurred. "Substantial meltdown" is the phrase used in 10CFR100.³ Melting of approximately 75% of the fuel was utilized by the ARSAP group as the starting point for fission product release estimates. This is considered to be a conservative estimate of fuel melting for a recovered accident. Individual accident sequence source terms, including unrecovered accidents, are to be calculated as part of the probabilistic risk assessment (PRA) in accordance with the ALWR Requirements Document and the PRA Key Assumptions and Groundrules (Chapter 1, Appendix A of the Requirements Document).

2.0 FISSION PRODUCT RELEASE TIMING

2.1 Introduction

The purpose of this section is to define more realistic timing than currently specified for the release of fission products from the fuel to the RCS and to the containment. Existing source term regulatory guidance includes the assumption that the entire source term is released instantaneously at the time of the initiating event [i.e., a large loss of coolant accident (LOCA)]. Although this assumption is consistent with other parts of the regulations (i.e., calculation of dose for two time periods following release, two hours and thirty days), it is physically impossible. Further, in the application of the source term as the design basis for various engineered safety systems, the assumption of an instantaneous release can result in distortions in the dose evaluation and less-than-optimum system designs.

An example illustrating the effect of the instantaneous release assumption is the design of containment spray additive systems for many existing PWRs. The concept of the system arises from the need to maintain a favorable partitioning of iodine between liquid and gas phases following release of iodine from the fuel. In past regulatory guidance, the assumption of an instantaneous release of large quantities of iodine to the containment vapor space at the time of the initiating event required actuation of the spray system in a matter of seconds, and fast delivery of relatively large quantities of sodium hydroxide to the containment. This requirement in turn resulted in added system complexity, potentially corrosive conditions as a result of system malfunctions, tightly controlled technical specifications on additive chemistry and system configuration, and rather costly clean-up from inadvertent operation of the system. If it is recognized that volatile iodine releases to the containment would take on the order of hours, the same objective of adjusting water chemistry for long-term retention of volatile iodine species in the liquid phase can be accomplished by the simpler approach of storing anhydrous pH adjusting chemicals in the containment sump.

2.2 Timing and Magnitude of Coolant and Gap Activity Release

Although release timing on the order of hours may be realistic for large fission product quantities resulting from fuel melting accidents, it is recognized that this would not be an appropriate criterion for the actuation time of systems designed to prevent releases of much lower magnitude such as the coolant and gap source terms. The actuation time for the isolation of containment purge valves is an example of a criterion which should be derived from timing of fission product release from the coolant or gap as opposed to from molten fuel. For applications of the source term release assumptions to mitigation systems, including actuation signals for such systems, it is important to note that any large fission product release from the fuel would be preceded by smaller releases of gaseous activity from the fuel gap, and, for loss-of-coolant accident scenarios, by the release of the activity circulating in the primary coolant. While these releases would occur sooner than a large fuel release, the curie inventories involved are smaller by perhaps two or more orders of magnitude. Table 2 gives a qualitative summary of the relationships of timing and magnitude for the major components of the fission product source term.

By comparing Table 2 to the existing regulatory source term it is apparent that regulatory guidance combines the largest fission product inventory with the earliest release timing. In a more realistic treatment of release timing, the large differences in the inventory available for release in each of the three components of the source term must be recognized. A potential treatment of release timing which recognizes these differences in inventory is shown in Table 3.

The "release timing" entry in Table 3 reflects the earliest time that any significant fraction of the release component can be expected to be released into the containment atmosphere for a full spectrum of accident sequences, including sequences involving fuel damage (i.e., substantial melting of the core). Although not reported here, passive plant work is being pursued to develop more precise magnitude and timing estimates for coolant and gap inventories.

TABLE 2. RELATIONSHIP OF RELEASE MAGNITUDE AND TIMING

	<u>Inventory</u>	<u>Release Timing</u>
Coolant Activity	very low	very early
Gap Activity	low	early
Fuel Activity	high	late

TABLE 3. POTENTIAL RELEASE TIMING MATRIX

<u>Source</u>	<u>Start of Release</u>
Coolant Activity	1 minute
Gap Activity	30 minutes
Fuel Activity	1 hour

A large LOCA results in the earliest release of coolant activity to the containment. If an essentially instantaneous large break is postulated, coolant activity would begin to be released in a matter of seconds, and, depending on the postulated size of the break, would be largely complete in a few minutes. The transport and mixing in the containment would take on the order of seconds to minutes, so that availability for release from the containment can be postulated to begin in about a minute for large breaks.

The release of gap activity requires the failure of the fuel cladding. It should be noted that the 30 minutes for the onset of gap activity release would not be negated by the failure of a few fuel rods during the early phases (e.g. blowdown) of an accident, as the activity in the gap of several fuel rods is of the order of the coolant activity. Failure of a significant fraction of the rods, which would result in the releases of the order of magnitude corresponding with the gap inventory of the core, would not occur sooner than about 30 minutes for any best estimate calculation.

As discussed in Section 1 the focus of this report is on large releases and the calculation of the corresponding offsite doses. While it is noted that the coolant and gap activity releases should be included in the design basis for certain safety systems, siting dose calculations should be based on the large fuel activity release. The remainder of this section will address the release timing of fuel activity. A more detailed ARSAP effort on ALWR coolant and gap activity is underway and a separate report will be prepared when this effort is complete.

2.3 Release Timing for Fuel Component of Source Term

The release of a significant fraction of the fission product inventory from the fuel matrix requires either long-term heating of the core following fuel failure, or substantial liquefaction of the fuel structure by dissolution or melting. Based on an examination of a spectrum of existing plant severe accident sequences as discussed in an ARSAP report on timing of fission product release,⁴ and taking into account ALWR features (such as no large pipes entering the BWR reactor vessel below the top of the core), the release of a significant fraction of the fuel inventory would not begin for about one hour or longer in the ALWR.

Once release begins, experimental results as well as the TMI-2 accident indicate that significant additional time is required to release the bulk of the volatile fission products. While analysis of the important sequence types will be necessary to define this timing, uniform release over 30 minutes has been used in the source term update for evaluating BWR pool scrubbing and PWR containment spray in order to obtain a meaningful release to containment and associated dose for the two hour exclusion area boundary dose calculation required in 10CFR100. Without the need to do a two hour dose calculation, the release period could be significantly longer than 30 minutes since realistic fission product release generally occurs over a much longer period.

3.0 MAGNITUDE OF FISSION PRODUCT RELEASE FROM FUEL INTO THE RCS

3.1 Introduction

In this section, fission product releases from the fuel to the RCS are estimated and justifications for these releases are provided. The starting point for the estimate of fuel releases is the ~ 75% fuel melt noted in Section 1.3 above. The 75% fuel melt is considered to be a conservative estimate of the extent of melting, and thus the fission product release from the fuel, for recovered sequences. For example, with the core in-vessel, radial heat losses to the sides of the reactor vessel and axial heat losses to the upper plenum and to water in the lower plenum will very likely prevent the core from progressing to ~75% meltdown. Further, the availability of diverse, active water addition systems increase the likelihood of accident recovery in the evolutionary plant. Finally, ALWR features for flooding of the reactor vessel cavity/lower drywell provide cooling of core debris and ex-vessel recovery. It is noted that about 45% of the core melted in the TMI-2 accident.

3.2 Proposed Release Magnitude

Justification for proposed release magnitude for noble gases, iodine and cesium, tellurium, and semi- and low volatile fission-products are provided in Sections 3.2.1 - 3.2.3. The suggested release magnitudes are tabulated in Section 3.2.4.

3.2.1 Noble Gases, Iodine, and Cesium

Analysis of fission product releases from the TMI-2 accident^{5,6,7,8} and from severe fuel damage experiments^{9,10,11,12,13,14,15,16} indicate that the releases of noble gases, iodine, and cesium are virtually identical and are closely related to the fraction of the fuel that becomes molten in the accident sequence. In the TMI-2 accident, about 45% of the core was molten and the releases of noble gases, iodine, and cesium were in the neighborhood of 55%.

Measurements of residual fission products in previously molten fuel indicate that up to ~10% of the original cesium inventory and somewhat less of

iodine can be retained by the formation of chemical species that are stable at high temperatures and/or geometries having low surface-to-volume ratios (see References 6 and 17). On the basis of these results, releases of 90% of iodine and cesium from molten fuel are proposed. No residual fission gases were found in molten fuel debris from TMI-2 (see Reference 5), so 100% release of noble gas from molten fuel is proposed.

The fractional release of fission products from the 25% of the fuel which does not melt should also be considered. The release of noble gases, iodine, and cesium increases with the extent of oxidation of the unmelted UO_2 fuel by steam during the heatup in an accident. In addition, fission product release occurs as a result of fuel pellet cracking during reflood. A release of 25-30% of noble gases, iodine, and cesium from unmelted fuel in a terminated accident appears to be a reasonable bound based on data from TMI-2 and the severe fuel damage tests conducted at the Power Burst Facility at INEL.

Fission product releases from fuel in the TMI-2 accident and in the severe fuel damage tests are presented in Tables 4 and 5. These data support a release of about 80% for noble gases (100% from melted fuel and 25-30% from unmelted fuel) and 75% for iodine and cesium (90% from melted fuel and 25-30% from unmelted fuel) given an accident with about 75% fuel melting.

3.2.2 Tellurium

Considerable study has resulted in the understanding that tellurium is released from the fuel at about the same rate as noble gases, iodine, and cesium, but it is largely retained by the surrounding metallic zircaloy cladding and is released during oxidation of the cladding.^{18,19} Tellurium has a chemical affinity for metallic zircaloy and most other metals. Oxidation of the cladding has the effect of increasing the concentration (and therefore the chemical activity) of tellurium in the remaining metallic zircaloy, thereby increasing the partial pressure of tellurium. When the local oxidation of zircaloy is equivalent to less than about 70% active clad conversion to ZrO_2 , the release rate of tellurium has been found to be 1/40 that of iodine and cesium, but equivalent to that of iodine and cesium when zircaloy oxidation exceeds 70%. The data in Tables 4 and 5 indicate that a value of 40% for

tellurium release bounds most conditions for oxidation and melting in recovered accidents.

3.2.3 Semi-Volatiles and Low Volatiles

The releases of strontium, barium, antimony, and ruthenium have been found to be quite low as demonstrated in Tables 4 and 5, and are bounded by a value of 1%. Barium and strontium exist as oxides within the UO_2 under accident conditions and have low volatilities (see Reference 15). Antimony and ruthenium are present as metals which are insoluble in the oxide fuel matrix and tend to separate from the fuel, concentrating with molten metallic debris (see Reference 17). Cerium, lanthanum, and the actinides (uranium, plutonium, americium, curium) are oxides with very low volatilities that are dissolved in the fuel matrix and thus are released to a very small extent (<0.01%) (see Reference 15).

3.2.4 Suggested Release Magnitudes

The proposed releases from fuel are listed in Table 6. Some of these proposed releases are larger than those in TID 14844 (I, Cs, Te), some are smaller (Xe, Kr, Ce, La, actinides [these include U, Pu, Am, and Cm]), and others are the same (Sr, Ba, Ru, Sb). It should be noted, however, that a direct comparison of these releases (other than noble gases) with the existing regulatory source term cannot be made since the TID 14844 releases are to containment, whereas the releases of Table 6 are to the RCS.

TABLE 4. RELEASES FROM THE CORE IN THE TMI-2 ACCIDENT

<u>Isotope</u>	<u>Fraction of Core Inventory Released</u>
^{85}Kr	0.54
^{129}I	0.55
^{137}Cs	0.55
^{132}Te	0.06
^{90}Sr	0.001 ^a
^{106}Ru	0.005
^{125}Sb	0.016
^{144}Ce	0.0001

- a. Leaching from damaged core after reflood increased Sr release to 0.032 two months after accident.

TABLE 5. FUEL RELEASE FRACTIONS FROM PBF SEVERE FUEL DAMAGE TESTS

<u>Element/ Exp. Cond.</u>	<u>SFD-ST</u>	<u>SFD 1-1</u>	<u>SFD 1-3</u>	<u>SFD 1-4</u>
Kr, Xe	0.50	0.026-0.093	0.08-0.19	0.23-0.44
I	0.51	0.12	0.18	0.26
Cs	0.32	0.09	0.18	0.44-0.56
Te	0.40	0.01	0.01-0.09	0.03
Ba	0.011	0.006	0.004	0.008
Sr	0.00002		0.00024	0.0088
Sb			0.00019	0.0013
Ru	0.0003	0.0002	0.00003	0.00007
Ce	0.000002	0.00009	0.00008	0.00013
Actinides			<0.0001	<0.00001
%Zr Oxidized	75	26	22	32
%Fuel Melted	15	16	18	18

TABLE 6. PROPOSED RELEASES FROM FUEL TO RCS

<u>Elemental Group</u>	<u>Release from Fuel (fraction of core inventory)</u>
Noble gases (Xe,Kr)	0.80
Iodine and Cesium	0.75
Tellurium	0.40
Semi-volatiles (Sr,Ba,Sb,Ru)	0.01
Low volatiles (Ce,La,Actinides)	0.0001

4.0 FISSION PRODUCT CHEMICAL FORMS

4.1 Introduction

The chemical form for iodine and cesium in the licensing source term should be determined by considering the chemical environment which each experiences after being released from the fuel. For more than a decade evidence has been accumulating from thermodynamic analyses, from in-pile and out-of-pile experimental programs, and from evaluation of TMI-2 accident data which show that CsI and CsOH will be the dominant chemical forms of iodine and cesium which undergo transport in LWR core damage accidents. Chemical and physical forms of other fission products are addressed briefly in Section 4.4.

4.2 RCS Speciation of Iodine and Cesium

At the high temperatures characteristic of core damage the iodine and cesium are assumed to escape from the fuel material as atomic species and enter the steam-hydrogen gas mixture flowing up through the core. As this mixture moves downstream and cools thermodynamic analyses predict that CsI and CsOH will be the stable end products.^{20,21} Since the core mass inventory of cesium is typically about ten times that of iodine, and the release rates from fuel for both are similar, the molar ratio of CsOH/CsI in the mixture should be about 10. The excess of CsOH helps protect CsI from thermal hydrolysis by steam²² and is also helpful against reactions with other vapor phase material which might be present such as boric acid.²³

As already noted the results of several experimental programs are in agreement with the above predictions. In the STEP tests²⁴ fission product iodine was frequently found to be collocated with fission product cesium on deposition coupons and aerosol collection samples. In addition deposit morphology was consistent with the presence of CsOH which would have been a liquid droplet aerosol at test conditions. The investigators concluded that CsI was the principal iodine containing species in the tests, and they also concluded that flow blockages in two of the tests probably had been caused by accumulation of viscous CsOH plus structural component aerosol material at constrictions in the downstream flow systems. In the SFD tests²⁵ the deposition patterns of Cs and I fission products were very similar and it was

concluded that the overall behavior of iodine in these tests was consistent with that predicted for CsI, but inconsistent with the assumption that the iodine was elemental or hydrogen iodide. CsOH was also identified as the dominant cesium form. In the LOFT FP-2 test the deposition pattern of fission product iodine indicated that it existed as an aerosol rather than a gas in the upper plenum.²⁶ Analysis of the test results indicates that AgI was probably the dominant chemical form of iodine in that particular experiment (i.e., low burnup fuel, low pressure RCS, and Ag-In-Cd control rod failure in the upper core region prior to fission product release). No evidence was found for volatile forms of iodine. In a series of out-of-pile fission product release experiments with high burnup fuel at ORNL²⁷ the investigators concluded from analysis of thermal gradient tube deposition profiles that CsI and CsOH were the dominant downstream iodine and cesium species for conditions which simulated LWR core damage conditions. Finally, measurement of the iodine speciation in the containment sump water from the TMI-2 accident followed by an analysis of how the species could have been produced concluded that fission product iodine entered the water primarily as iodide and not as elemental iodine (see References 6 and 8).

Boric acid is known to react with CsI to produce HI. Although quantification of the HI from this effect is difficult, it is judged that the amount of HI produced which escapes the RCS to the containment would be very small due to the excess of CsOH noted above, the need for intimate mixing of the vapor phases of boric acid and CsI (see Reference 23) and the tendency of HI to react chemically with metallic structures and aerosols. The LOFT FP-2 (see Reference 26) experiment supports this judgment since this experiment was borated, and little, if any, evidence of CsBO₂ was found during pre-reflood. Further, as noted above, no evidence was found for volatile forms of iodine in LOFT FP-2.

Although CsI has not been explicitly measured as an iodine species in work done to date, the accumulated experimental evidence and accident experience strongly support the position that particulate iodine (CsI or possibly some AgI) will be the dominant iodine species released to containment from the RCS in an LWR core damage accident. Likewise, theory and experimental evidence (while indirect) are in agreement that the dominant form for fission product cesium will be CsOH.

4.3 Containment Speciation of Iodine and Cesium

At containment temperatures which are predicted to occur in an ALWR core damage accident (i.e., at or near saturation temperature) the CsI and CsOH released from the RCS will exist in aerosol form and will participate in all ongoing aerosol removal processes. Any AgI would behave similarly. The CsOH should react with any atmospheric CO_2 that is present to rather quickly form Cs_2CO_3 which would then more slowly convert to CsHCO_3 . However, aerosol removal should not be strongly affected and these processes will effectively result in the steady buildup of fission product iodine and cesium in the water reservoirs present within the containment. Each of the above compounds are quite water soluble and dissociate in solution to yield non-volatile ionic species. Thus iodine would exist as I^- ions and cesium as Cs^+ ions.¹

However, in the presence of radiation levels, such as would be expected in a core damage accident, recent research has shown that aqueous I^- may be readily oxidized to I_2 and this can lead to relatively high steady-state gas phase iodine concentrations in the vapor space above the water reservoirs.^{28,29,30} The amount of I_2 formed is a strong inverse function of solution pH; in fact, at a pH of 9 the radiolytic effect is virtually eliminated[28]. While most of these data were obtained at room temperature the strong pH dependence should persist at higher temperatures. The importance of controlling pH in this situation is clear and so it is assumed here that measures will be taken in ALWRs to assure that the pH of the containment water is maintained in an alkaline state for the accident duration.

In addition to water pool radiolysis, three other containment processes that could potentially generate some I_2 are hydrogen combustion events, evaporation to dryness of shallow water puddles, and radiolysis of acidic droplets which have absorbed HI (from boric acid volatilization) which may have been released from the RCS. While limited experimental data and uncertainties in accident progression make highly accurate quantification difficult, the I_2 yields from all of these should be small. CsI that is in

1. Silver iodide is only slightly soluble in water and would not dissociate to any significant extent.

solution in a water pool would not be affected by a hydrogen deflagration. Thus, in order to produce a large effect, an energetic hydrogen deflagration would have to occur when most of the CsI aerosol is still suspended in the containment atmosphere.³¹ This is very unlikely for a recovered accident where the conditions for significant hydrogen deflagration (i.e., high hydrogen concentration, low steam concentration) would not occur until late in the accident when little suspended CsI remains. Further, in the saturated atmosphere expected to exist in containment, the condensed steam appears to be protective even if a burn occurs. Similarly the fraction of total containment water containing iodide that might undergo evaporation to dryness during an accident should be very small and so generation of I_2 by this process³² should also be small. Finally, the I_2 resulting from radiolysis of I^- in acidic droplets should be very small since HI is very hygroscopic and will be readily neutralized by interaction with alkaline aerosol.

4.4 Numerical Estimates of Iodine and Cesium Chemical Forms

Accident consequence assessment requires numerical input regarding containment airborne forms. In the case of iodine a high fraction should be CsI. The three containment processes noted above could each produce a small amount of I_2 . As a first estimate these three sources of I_2 are considered roughly equal and a reasonable value for each is judged to be equivalent to 1% of the total iodine in containment. Thus the total I_2 yield would be 3%. The balance would be CsI, except that the presence of I_2 now means that the generation of organic iodide (primarily CH_3I) must be considered. It is generally recognized that thermal and/or radiolytic reactions between I_2 and a wide range of organic substances which can be present in containment vessels are responsible for the appearance of organic iodides in these systems.^{33,34,35} Measured yields depend on a variety of parameters which include I_2 concentration, temperature, radiation dose, type of organic, and geometry effects among others. No completely satisfactory predictive method has evolved but an empirical procedure (see Reference 33) was devised some time ago which has had considerable use. This procedure which tends to overpredict organic iodide yields would indicate conversion of roughly 5% of the I_2 in the present case into organic species. This amount of organic iodide would then correspond to about 0.15% of the total iodine in the containment.

In summary, on the basis of the evidence that has been cited and the restrictions that have been stated, the containment speciation proposed for fission product iodine and cesium is as follows:

Cesium - Airborne cesium will exist entirely as a particulate aerosol composed of mixed salts which are highly soluble in water. It will not volatilize from solution.

Iodine - Airborne iodine will exist as a mixture of three species: 97% particulate, primarily CsI which is a highly soluble particulate aerosol, 2.85% as I_2 which is a moderately soluble vapor, and 0.15% as CH_3I which is a slightly soluble gas.²

4.5 Forms of Other Fission Products in Containment

The other fission products considered here include tellurium, the semi-volatiles (barium and strontium), and the low volatiles (lanthanides, actinides, noble metals, etc.). The relatively small portions of the core inventories which might reach containment for these materials are expected to consist of a variety of chemical species including salts, hydroxides, oxides, intermetallics, etc. which would exist in aerosol form at containment conditions. Some of the compounds are water soluble while others are not but none would be expected to volatilize from alkaline water pools or sumps.

²

It is recognized that the numbers for elemental and organic iodine may be more precise than warranted; however, the total is constrained to sum to 100%, and further adjustment or rounding off is left to the process of formally implementing the source term update.

5.0 RCS RETENTION

5.1 Introduction

Fission products released from the fuel during core damage events will be affected by physical and chemical processes during transport through the RCS which in turn affect the retention of aerosols in the RCS. Retention of the fission product aerosols in the RCS has an important effect on the source term, so evaluation of these processes must be included.

The amount of retention in the RCS depends not only on the design of the plant but also on the details of the accident sequence being considered. The NRC and nuclear industry have developed computer codes (e.g. TRAP-MELT and MAAP) which predict the extent of deposition in the RCS for various accident sequences and have undertaken experimental programs for the purpose of validating these methods.

As noted above in Section 4, thermodynamic analysis and experimental evidence indicate that iodine, cesium, and less volatile radionuclides released from the fuel during core damage accidents in LWRs will behave primarily as aerosols. The aerosols will experience forces that deposit substantial fractions of the material on RCS surfaces or in water reservoirs. The evidence from the TMI-2 accident³⁶ indicates that for sequences in which the transport pathway is partially water filled, iodine and cesium will follow the liquid streams and be retained primarily as soluble species in solution.

The evidence presented in Section 4 further suggests that the dominant chemical forms of the iodine and cesium are CsI (and possibly some AgI) and CsOH. These dominant chemical forms will exhibit condensed phase behavior at the temperatures expected in the RCS for a terminated accident in-vessel.

5.2 Experimental Evidence on RCS Retention

Experimental evidence of aerosol RCS retention processes is provided by the LACE^{37,38} and Marviken³⁹ aerosol transport tests as well as by the SFD 1-4 test (see Reference 13) and the LOFT FP-2 test (see Reference 26). Table 7 summarizes the results. Aerosol retention in the piping system of about 80%

TABLE 7. SUMMARY OF EXPERIMENT RETENTION FRACTIONS (% OF SOURCE)

Test	Species	DEPOSITION	
		Close to Fuel (Source)	Total Piping
LACE LA3A	CsOH/MnO=.21	26	77
LA3B	CsOH/MnO=.13	15	51
LA3C	CsOH/MnO=.61	46	83
LA1	CsOH/MnO=.43	--	99
Marviken	--	--	~74
SFD 1-4	Iodine	10	95
	Cesium	30	95
LOFT FP-2	Iodine	66	70
	Cesium	60	71

was measured in LACE tests LA3A and LA3C which had soluble/nonsoluble aerosol ratios on the order of that expected from core damage accidents. Test LA3B had a lower retention, probably due to a very low soluble/nonsoluble aerosol ratio. The Marviken tests used prototypic core materials and found ~74% retention in the RCS. These large retention fractions are representative of that expected when a piping system is included in the transport path, and deposition at bends due to particle impaction is a dominant removal mechanism. Retention fractions of the order of 25 to 50 percent were noted for the first few meters of piping.

The SFD 1-4 test measured fission product deposition on surfaces downstream of the damaged fuel region. Large fractions of iodine and cesium (up to 30%) were found to deposit close to the fuel, although some material was able to migrate long distances (~20m) before being deposited. Total system retention was 95%.

The LOFT FP-2 test simulated a LOCA without emergency coolant makeup in which fission products were transported from the RCS through a long LPIS line. During the pre-reflood phase of the test 2-3% of the volatile fission products were released from the fuel. Approximately 2/3 of the released iodine and 1/2 of the cesium were deposited in the reactor vessel and hot leg pipe, and nearly 75% of this material was retained in combined RCS piping and the LPIS line. Because these experiments were performed with real fuel and control rod materials within a prototypic geometry, the fission product deposition behavior was controlled by aerosol processes, and is expected to be representative of RCS deposition behavior in an actual plant.

5.3 Analytical Results on RCS Retention

The experimental evidence is quite supportive of the argument that large fractions of iodine, cesium and less volatile radionuclides will deposit on system surfaces during transport through the RCS. However, as noted in Section 5.1 the amount of RCS retention is dependent on the design details of the transport pathway and the thermal-hydraulics of the accident sequences. In support of NUREG-1150,⁴⁰ the NRC's TRAP-MELT code⁴¹ (one of the modules of the Source Term Code Package) was used to estimate the amount of RCS retention that can be expected for a variety of accident sequences in modern, operating PWRs and BWRs.⁴² The predicted retention factors for aerosols in the RCS range from approximately 15 percent to 85 percent. The lowest values are associated with large, hot-leg pipe break accidents in PWRs and low to intermediate pressure sequences in BWRs in which core uncover occurs early (about one hour after shutdown). Four considerations must be factored into the evaluation of these computer code results relative to ALWRs: revaporization in recovered sequences, improved understanding of the likelihood of primary pipe breaks, limitations in the computer code, and differences in ALWR design vs. operating plants. These considerations, as discussed below, suggest that the low values of RCS retention are not applicable to the ALWR.

The retention factors predicted by TRAP-MELT for iodine and cesium tend to be less than those predicted for lower volatility aerosols because of the potential for revaporization of deposited iodine and cesium prior to vessel failure. However, since much of the iodine and cesium revaporization predicted by TRAP-MELT occurs late in the in-vessel phase of unrecovered core damage sequences, it is unlikely that there would be significant differences between iodine, cesium, and bulk aerosol retention in recovered sequences.

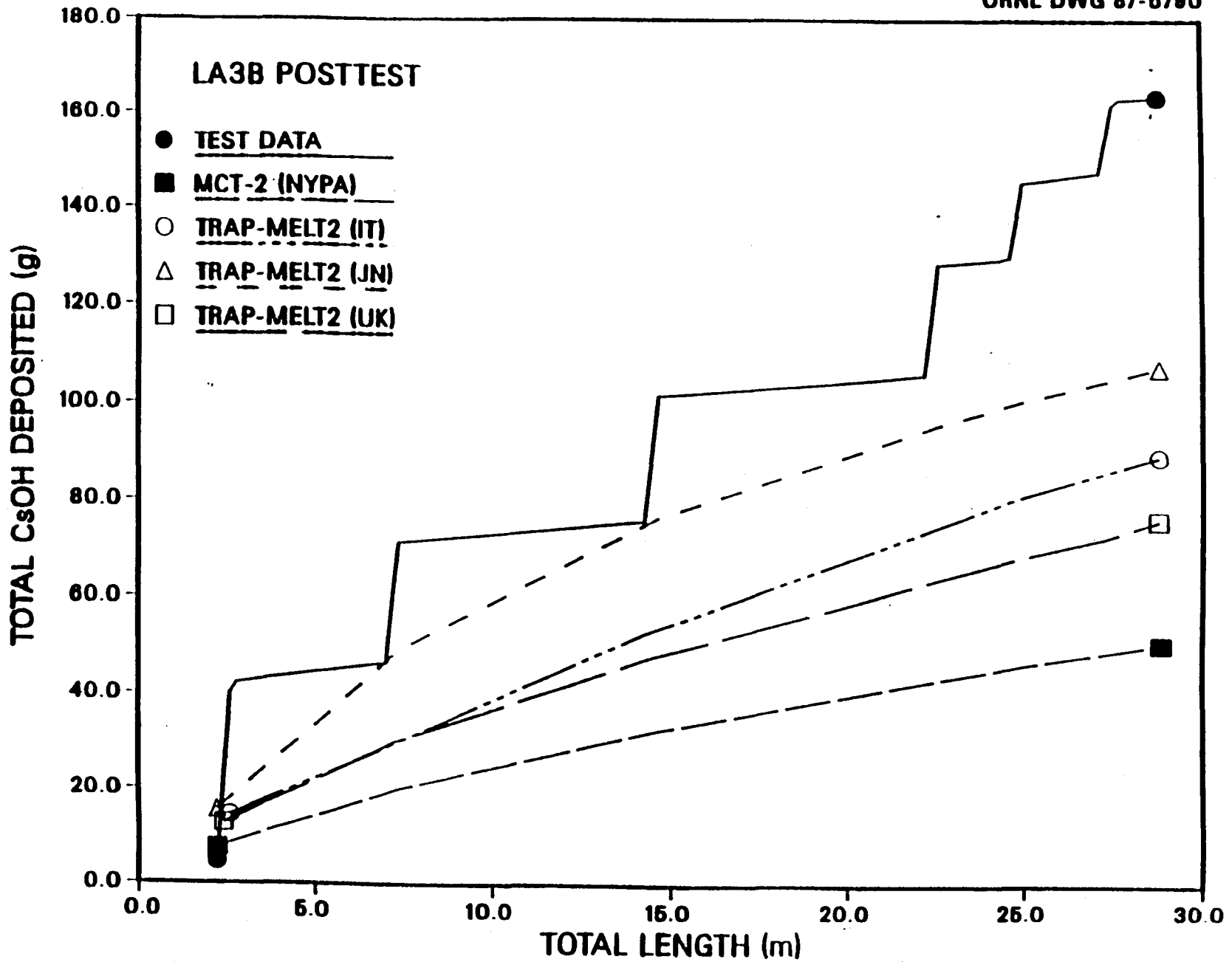
Extensive experimentation and PRA analysis have shown that large RCS pipe break initiated core damage sequences are very low in probability ($\leq 10^{-7}$ per year). Such sequences are reduced even further in likelihood by application of leak-before-break technology. Extensive investigations of the fracture mechanics of piping provide confidence that a leak in primary system piping would precede ruptures, thus allowing the plant to be shut down and the RCS depressurized before a break could occur. The NRC has recently issued an

amendment to General Design Criterion⁴³ which acknowledges the need to address application of leak-before-break to requirements other than dynamic effects of pipe rupture. This further reduction in likelihood of an already very low probability core damage sequence suggests that significant size pipe breaks located close to the reactor vessel need not be part of the basis for determining RCS retention for ALWR source term estimates.

The version of TRAP-MELT used in the Source Term Code Package is recognized to underpredict aerosol retention within the RCS because of unmodeled phenomena. In particular, this version does not model the effect of bends on particle deposition, a process that has been shown to be important in experiments as noted in Section 5.2. Figure 1 illustrates a post-test comparison of deposition measured in test LA3B versus predictions with versions of TRAP-MELT that do not contain models for predicting deposition in bends. Figure 2 shows the same test results compared with calculations of codes which do model bend deposition. The rapidly rising sections of the experimental curve represent regions of high deposition at bends. The TRAP-MELT 2.2 code, which was quite successful in predicting deposition in the LACE LA3 test series, is an experimental version of the code which incorporates a turbulent deposition model for treating aerosol deposition at bends. On the basis of the Figure 1, Figure 2 comparison, it is evident that RCS retention estimated by codes like TRAP-MELT 2.0 (used in NUREG-1150) will be underpredicted.

In the Source Term Code Package analyses, particle agglomeration and sedimentation are found to be the principal aerosol mechanisms leading to retention in the RCS. As a result the residence time within the RCS is the dominant parameter affecting retention. Thus accidents with low fission product residence times, i.e., those that result in high flow rates, have low predicted retention factors. Turbulent deposition at bends does not have this same dependence on residence time, however, and can be expected to contribute substantially in accident scenarios in which sedimentation is not large.

Figure 1
POST-TEST COMPARISON OF LA3B DEPOSITION
VS. TRAP-MELT PREDICTIONS WITHOUT EFFECT OF BENDS



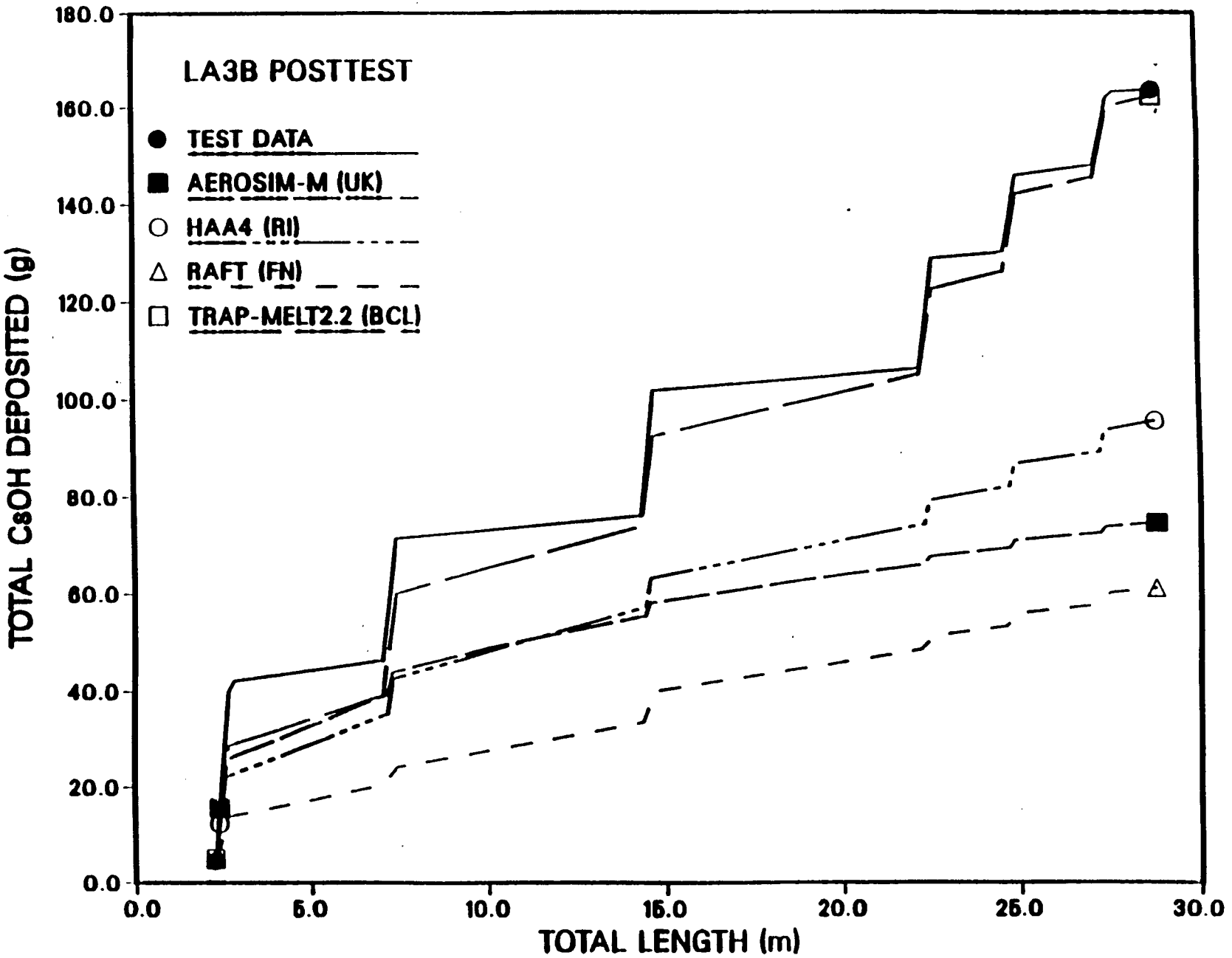


Figure 2
POST-TEST COMPARISON OF LA3B DEPOSITION
VS. TRAP-MELT PREDICTIONS WITH EFFECT OF BENDS

An uncertainty analysis was performed as part of NUREG-1150 in which ranges were determined for uncertain parameters such as the RCS retention fraction by polling source term experts. In their evaluations the experts recognized the limitations of existing RCS deposition codes. Table 8 shows the median values obtained by evaluating the expert responses for different types of accident scenarios. The lowest values are again associated with low pressure accidents and accidents involving early core melt and revaporization, but these values are higher than the TRAP-MELT predictions and thus appear to have accounted for unmodeled retention mechanisms. It is noted that the expert opinions solicited for NUREG-1150 were for unrecovered severe accidents for existing plants.

A final point regarding the applicability of the TRAP-MELT results and NUREG-1150 estimates is that both the PWR and BWR evolutionary designs have depressurization systems which would be used to depressurize the RCS in the event of a core damage accident. Although RCS retention in these sequences would be similar to that in large pipe break accidents, the pathway to containment is via a large pool of water (suppression pool or in-containment refueling water storage tank) where substantial retention of aerosols would occur. Also, ALWRs tend to have slightly larger RCS volume to power level ratios, which leads to delayed uncover of the core and longer residence times during the period of release.

5.4 Summary of Estimated RCS Retention

A value of 70 percent is suggested for RCS retention for all aerosols. The amount of RCS retention that would occur in a severe accident would depend on the timing and thermal-hydraulic conditions of the particular accident sequence. The 70 percent value is considered a lower bound to the best estimate retention over a range of accident sequences based on the following:

- Experimental evidence indicating 70% or higher for aerosol retention in vapor pathway piping systems where the aerosol material and the controlling thermal-hydraulic conditions are similar to that of actual reactors.

Table 8
NUREG 1150 EXPERT ELICITATION MEDIAN RETENTION FACTORS

Case	Conditions	Percent Retention		
		Iodine	Cesium	Low Volatility Aerosols
PWR1	Setpoint pressure	91	96	97
PWR2/3	High and intermediate pressure	59	71	76
PWR4	Low pressure	48	60	66
BWR1	High pressure, early melt	91	97	97
BWR2	Low pressure, early melt	59	70	74
BWR3	High pressure, de-layed melt	72	75	92

- Experimental evidence and TMI-2 evidence indicating nearly complete aerosol retention in liquid pathways.
- The extremely low likelihood of a core damage accident initiated by large, close to vessel pipe breaks. Extensive investigation of the fracture mechanics of piping provide confidence that leaks in primary system piping would precede ruptures and would be detectable, allowing the plant to be shutdown before a break could occur. This will significantly reduce the already very low frequencies of large LOCA initiated severe accidents obtained in PRAs.
- Extrapolation of analytical results and NUREG-1150 expert judgement to account for the extremely low likelihood of significant size, close to vessel pipe breaks and for reduced revaporization of volatile species prior to vessel meltthrough, which would be expected for a recovered accident.
- ALWR design features which would tend to increase aerosol retention beyond that expected for existing LWRs, e.g., in-containment refueling water storage tank, larger RCS volume.

6.0 FISSION PRODUCT REMOVAL IN CONTAINMENT

6.1 Introduction

Previous sections of this report have dealt with the release of fission products from the fuel and their transport through the RCS, including their chemical form. It has been established that the dominant physical form of the fission products (other than the noble gases and a small fraction of the iodine) upon their release to containment will be as particulate. The size distribution will be in the aerosol range; i.e., less than 0.1 mm (100 μm) in diameter. From the release fractions and containment free volume, it can be estimated that concentrations (including nonradioactive structural aerosols) will be several g/m^3 in the containment atmosphere.

The current regulatory approach to spray removal of fission products suspended in the containment atmosphere is largely oriented toward elemental iodine since this is assumed to be the primary chemical form of iodine and since no other elements (besides noble gases) are assumed to be released to the containment atmosphere. The treatment of particulate iodine is very conservative in current practice (see Appendix A, Section A.2.2). This has been acceptable because particulate was viewed as a minor component of the airborne release. With particulate now being viewed as the primary chemical form of iodine and with other fission product aerosols being considered, the regulatory approach for spray removal of aerosol from containment needs to be reexamined.

In BWRs, regulatory credit for suppression pool scrubbing of all fission products released to the drywell is allowed in Standard Review Plan (SRP) Section 6.5.5. With fission product release timing now being delayed as discussed in Section 2 above, the SRP 6.5.5 model needs to be updated.

6.2 Fission Product Removal in PWR Containments

The alternative to the current regulatory approach which is presented here applies available, mechanistic containment aerosol spray removal models in a bounding manner to the ALWR. This provides a more physically realistic treatment of aerosol removal mechanisms for the licensing source term.

In a PWR fission product aerosols escaping from the RCS in an accident will enter the containment atmosphere wherein sedimentation, diffusiophoresis, and spray removal depletion mechanisms will be operable as explained in Appendix A. Of these, spray removal is by far the most important and is the only removal mechanism being considered at this time in the evolutionary PWR licensing source term update. PWR containments are generally free of internal structures above the operating floor (with the exception of the polar crane and its supports), but are fairly well compartmented below the operating floor. In general, it is the region above the operating floor that is sprayed, and the highly compartmented region below the operating floor is normally unsprayed. In evaluating fission product removal in a PWR, it is important to know how much of the containment is sprayed and how much is unsprayed, and the degree to which the two regions mix.

6.2.1 Aerosol Removal in the Sprayed Region

In order to quantify the effectiveness of sprays for the evolutionary PWR, an analysis was performed for EPRI⁴⁴, Case C assuming the following:

- Core power of 3425 Mw(t).
- Large LOCA.
- Containment free volume = 3.5 E6 ft^3 , 70 percent sprayed.
- Spray flowrate = 3130 gpm.
- Characteristic spray droplet radius = $150 \text{ }\mu\text{m}$.
- Fission product release at a uniform rate over twenty minutes, beginning at $t = 20$ minutes (defined as $t = 0$ for dose calculation); total mass injected = 420 kg.
- Hydrogen production equivalent to 75 percent metal water reaction with a hydrogen burn initiated at the end of the fission product release (H_2 concentration = 5.5%).
- Hygroscopic treatment of CsOH.

The Reference 44 study was completed in January 1988, and several of the plant design and source term related inputs to that study have changed or been refined since that time. The changes include:

- Intermediate size as opposed to a large LOCA for source term (large LOCA still being used for ECCS and containment design).
- 75% of containment free volume above operating deck sprayed.
- Fission product release over 30 minutes beginning at one hour (and a corresponding delay in the hydrogen burn to the end of fission product release).
- Higher total mass injected due to changes in RCS retention.

Of these changes, the only notable effect on spray λ is to assume that the decrease in λ associated with the hydrogen burn occurs at 30 minutes rather than 20 minutes. Table 9 reflects a conservative adjustment of the data (calculated with NAUA⁴⁵ as described in Reference 44 to reflect this timing shift, and a conservative approximation of the Table 9 data is as follows:

- $\lambda = 100/\text{hr}$ for first 10 minutes of release (until $t = 0.17$ hours)
- $\lambda = 50/\text{hr}$ for next 30 minutes, including the last 20 minutes of release (until $t = 0.67$ hours)
- $\lambda = 5/\text{hr}$ from $t = 0.67$ hours until $t = 2$ hours
- $\lambda = 1/\text{hr}$ from $t = 2$ hours to $t = 24$ hours (removal need not be considered beyond 24 hours)

TABLE 9. SPRAY COEFFICIENT (λ) FOR THE SPRAYED REGION

<u>TIME (hr)</u>	<u>λ (hr⁻¹)</u>
0.0100	300.0
0.0139	265.0
0.0209	202.5
0.0278	140.0
0.0417	122.3
0.0556	104.6
0.0695	104.3
0.0833	104.0
0.0972	104.0
0.1111	104.0
0.1389	83.9
0.1667	63.7
0.1945	63.6
0.2222	63.4
0.2778	50.5
0.5000	18.8
0.6667	22.0
0.8333	5.7
1.0000	3.4
1.2500	2.9
1.5000	2.4
1.7500	1.9
2.0000	1.4

These values are considered to be appropriate for a release duration of 30 minutes and for containment spray systems designed in compliance with SRP 6.5.2 and for ratios of volumetric flowrate to sprayed region volume exceeding 0.01/hr.

These values of λ are substantially greater during the first half hour than those calculated using the SRP expression given in Section A.2.2 (approximately 7.5/hr for the removal of the first 98%, requiring about one half hour using the plant data listed above) and are moderately higher than the SRP values (0.75/hr from the SRP expression for the last 2%) at later times. It is only necessary to credit aerosol removal until the fraction of iodine airborne in aerosol form becomes much less than the organic fraction; at that point, the impact on consequence analysis will no longer be important. This will occur at times much less than 24 hours after the start of release.

6.2.2 Gaseous Iodine Removal in the Sprayed Region

Gaseous iodine removal calculations will conform to current regulatory practice as established in SRP 6.5.2. Typical values for elemental iodine removal coefficients with sprays are comparable to those given above for aerosol removal. Organic iodine (like the noble gases) is not considered to be removed from the containment atmosphere except by leakage to the environment and radioactive decay.

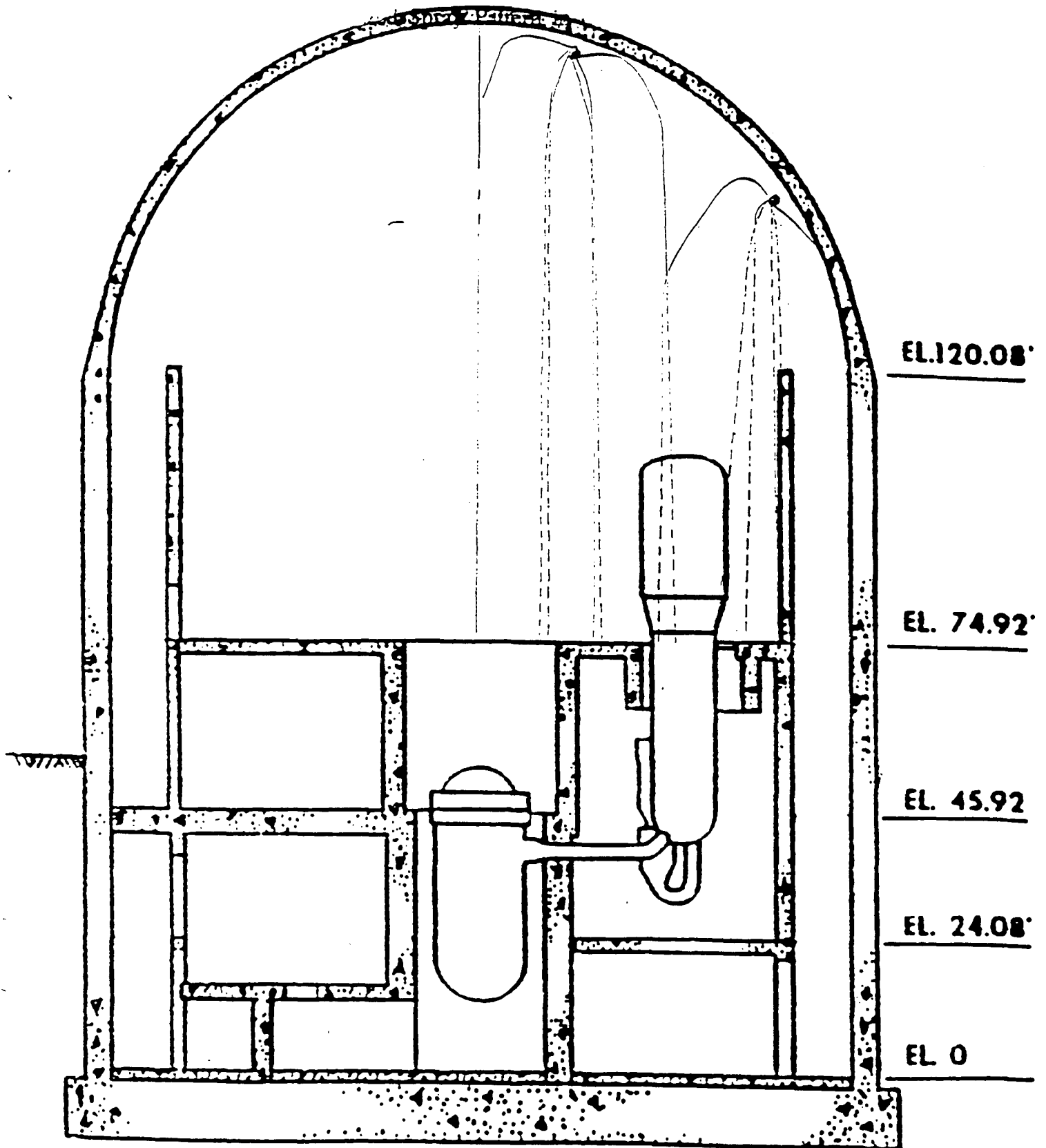
6.2.3 Removal in the Unsprayed Region

In practice, both sedimentation and diffusiophoresis (discussed in Appendix A, Sections A.1.2 and A.1.4, respectively) can be ignored in the calculation of fission product removal from containment compartments which make up the unsprayed region. This is because the most important effect is the potential for mixing with the sprayed region which is discussed in the following section.

6.2.4 Containment Mixing

6.2.4.1 Mixing Within the Sprayed Region. Figure 3 shows a typical, idealized spray pattern within a containment. The pattern is created by

Figure 3
IDEALIZED SPRAY PATTERN IN PWR CONTAINMENT

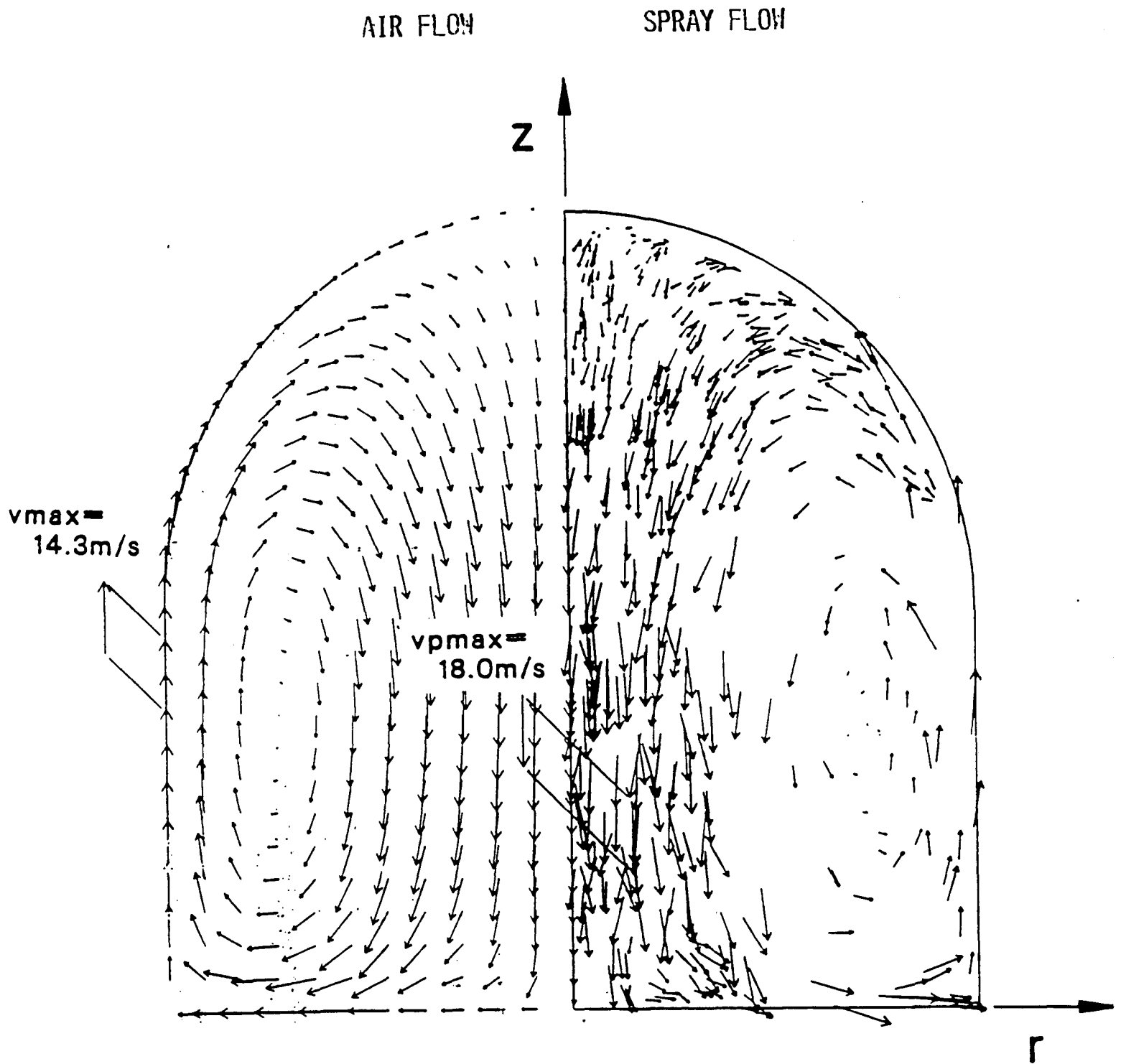


overlaying the spray patterns for individual nozzles with different locations and orientations. The individual nozzle spray pattern data are obtained from manufacturer test reports where nozzles have been tested individually, with a single nozzle spraying into a large chamber at various orientations and inlet pressures. Even though these spray patterns are adjusted for the effects of containment pressurization (increased atmosphere density and drag), there is no correction for the significant and sustained momentum exchange between the spray and the containment atmosphere.

A study of the effects of this momentum exchange (between the containment sprays and the closed containment atmosphere) was performed by Sandia⁴⁶ for the purpose of determining if induced air currents in an ice condenser containment would be sufficient to adversely affect igniter performance. The CONCHAS-SPRAY computer code was used in this study.

The baseline case for this study was a "clean" containment, i.e., one with no obstructions above the operating floor (only the region above the operating floor was modeled). The spray flowrate modeled was 2850 gpm. The results for the air flow and the spray droplet flow are shown on the left- and right-hand sides of Figure 4, respectively. Qualitatively, comparing Figures 3 and 4, it is evident that the real behavior of the sprays in the containment atmosphere will differ substantially from the idealized picture, and that the mixing in the sprayed region will be more intense than the idealized picture would indicate. The maximum air velocity for the baseline case was almost 50 fps; the maximum droplet velocity 60 fps. Relative velocities between the droplets and the recirculating containment atmosphere remained high. These observations tend to limit concerns with respect to spray "coverage" within so-called "sprayed" region of the containment.

Figure 4
REALISTIC SPRAY FLOW CONSIDERING MOMENTUM EXCHANGE
BETWEEN SPRAY AND AIR



6.2.4.2 Mixing Between the Sprayed and Unsprayed Regions. For purposes of this paper, a mixing rate between the sprayed and unsprayed regions is quantified by a simple model which depends only on the cooldown rate in the sprayed region and the buoyancy driven flow that results. This estimate is conservative in that it does not take credit for other important mixing mechanisms which would affect the unsprayed region including the momentum exchange effect discussed above, flow from the unsprayed region to the sprayed region due to steam condensation, and the effect of heat sources in the unsprayed region (i.e., heat sinks which become heat sources during the cooldown).

Assuming a containment that is initially well mixed (consistent with the assumption of a uniform distribution of fission products, including the compartments making up the unsprayed region) and with a cooldown rate in the sprayed region of dT/dt , a mixing rate, λ (expressed as a multiple of the unsprayed region volume per unit time), and a temperature difference between the sprayed and unsprayed region, δT , will have the following steady-state relationship:

$$\lambda = dT/dt/\delta T$$

and the steady-state condition (where the cooldown rate in the unsprayed region is also dT/dt) will be approached fairly readily (i.e., a quasi-steady model is acceptable). The symbol " λ " is used in this context because the mixing rate becomes essentially a removal constant for airborne material in the unsprayed volume as long as the concentration of the return flow is substantially less than that of the unsprayed volume, i.e.,

$$x = x_0 e^{-\lambda t}$$

where x is the airborne concentration as a function of time and x_0 is its initial value.

The other relationship that defines the two unknowns λ and δT is the one describing the buoyancy driven flow through some limiting effective flow area, $A/SqRtK$, with a driving head, H , head loss coefficient, K , and an unsprayed region volume, V_u . This relationship is as follows:

$$\lambda = A/\text{SqRt}K * \text{SqRt}(2g * H * \delta T / T_{\text{bar}}) / V_u$$

where g is the gravitational acceleration and T_{bar} is the average system temperature. For a typical PWR containment of current design $A/\text{Root}K = 1000 \text{ ft}^2$, $H = 25 \text{ ft}$ and $V_u = 500,000 \text{ ft}^3$ (based on a multi-node containment model),⁴⁷ and for the sprayed region cooldown rates observed in the analysis described in Section 6.2.1 ($dT/dt = 30^\circ\text{F/hr}$ for the first hour after the start of release and 10°F/hr for the second hour after the start of release), the mixing rate would be approximately 15 unsprayed volumes/hr for the first hour and approximately 10 unsprayed volumes/hr for the second hour. It was pointed out in reference 44 that mixing rates greater than 10 unsprayed volumes/hr have very little impact on dose reduction; accordingly, this is the maximum that will be suggested for use. For times greater than two hours beyond the start of release a mixing rate of two unsprayed volumes per hour (that permitted in SRP 6.5.2 without further justification by the user) can be assumed.

6.3 Fission Product Removal in BWR Containments

Probabilistic risk analyses of BWR plants indicate that, because of the diversity of water addition systems available in BWRs, pipe break accidents are much less likely to lead to fuel damage than transient types of accident initiators. In those transient sequences in which the flow path is through the safety/relief line and the sparger system to the suppression pool, the decontamination factor of the pool would be expected to be quite large (1,000 or larger per Appendix A, Section A.2.1). Because of the high effectiveness of pool scrubbing, the release to the environment from containment leakage would be quite small for these scenarios. Larger releases to the environment would be obtained for less likely accidents in which the release from the vessel is directly to the drywell as in a steamline break accident. The limiting case is a break in a steamline inside the drywell with initial failure of the emergency core cooling system. In this case, fission products released from the RCS flow completely to the drywell, as opposed to order scenarios in which the release is only partially to the drywell. The limiting accidents for containment leakage in the evolutionary BWR are those in which the release from the vessel is completely to the drywell, and this type of accident is assumed for purposes of updating the BWR source term.

As noted in Section 6.1 above, the existing regulatory guidance for pool scrubbing must be re-examined since the instantaneous fission product release no longer applies. In order to avoid the calculation of a variety of severe accident scenarios, the following approach was used for calculating the behavior of aerosols in the containment. The release to containment was assumed to begin at one hour following shutdown and to occur over a 30 minute period. During this 30 minute period, the average concentration of radionuclides in the drywell available for release is 0.4 times the total quantity released from the vessel. During the next one and one-half hours the amount available for release is 0.01 times the total quantity released from the vessel. The bases for these numbers used is discussed briefly below and in somewhat more detail in Appendix A, Section A.2.1.

The rationale for this approach involves the consideration of the amount of steam (or hydrogen) that must be produced in the process of uncovering and overheating the core and in the subsequent quenching of the core during recovery. The specific condition represented involves an extended period of core heatup (30 minutes) without any water addition. Any scenarios in which water is added sooner will tend to result in more rapid sweepout of the drywell contents to the suppression pool. It is assumed that at the completion of the 30 minute period of fission product release sufficient water is added to the vessel to at least prevent further core degradation (but not necessarily quench the core). The 0.4 release function is arrived at by assuming that the fission product release occurs uniformly over the 30 minute release period, resulting in an average airborne concentration of about 0.4 of the total release to the drywell.

For the remaining 90 minutes, as indicated in Appendix A, Section A.2.1, even the boiloff associated with decay heat alone is sufficient to rapidly sweep the drywell contents to the wetwell. Thus, all of the fission product source term from the vessel can be assumed to have been subjected to pool scrubbing after this time.

The time-integrated pool decontamination factor for this 90 minute period (i.e., the period which starts with rapid flow to the drywell) is expected to be 100 or greater. A number of factors will affect pool scrubbing, the most important of which are aerosol size and condensible/non-

condensable gas flow ratio. As discussed in Appendix A, Section A.2.1, consideration of these factors and the boundary conditions which encompass a range of accident scenarios suggest a pool DF of 100. Pool bypass factor will need to be applied to this DF to arrive at an overall decontamination for the 90 minute period .

7.0 SUMMARY OF PROPOSED LICENSING DESIGN BASIS SOURCE TERM FOR THE EVOLUTIONARY ALWR

Combining the results of Sections 2 through 6 yields a licensing source term update which is defined in Table 10. Also shown in the table is a comparison with the existing regulatory source term derived from TID 14844, Regulatory Guides 1.3, 1.4, and the SRPs. The ALWR source term update is expressed in the same form as the existing regulatory source term. The release to containment was obtained by multiplying the fuel release fraction by an RCS escape fraction. Table 11 defines this calculation.

It is recognized by the ALWR Program that the source term update described above could be taken further in the direction of a physically-based source term. As noted in Section 1, schedule and resource constraints have prevented doing this for the evolutionary plant, although such an effort is underway for the passive plant. Despite the schedule and resources constraints, the progress report presented here is considered to provide a useful update of the present regulatory source term and to provide a more rational basis for evolutionary plant accident mitigation system design.

TABLE 10. SUMMARY OF EVOLUTIONARY ALWR LICENSING SOURCE TERM UPDATE

	<u>Source Term Update</u>	<u>Existing Regulatory Source Term</u>
Release Timing	Release at uniform rate over 30 minute period beginning at 60 minutes after initiating event	Instantaneous at time of initiating event
Release Magnitude to Containment Atmosphere		
• Nobles	80%	100%
• Iodine	22.5%	25% ⁽¹⁾
• Cesium	22.5%	1% (to sump)
• Tellurium	12%	1% (to sump)
• Ba, Sr, Ru	0.3%	1% (to sump)
• Remainder	0.003%	1% (to sump)
Chemical Form in Containment		
• Iodine	2.85% elemental 97% particulate 0.15% organic	91% elemental 5% particulate 4% organic
• Cesium	100% particulate	Not specified
• Tellurium and Remaining Semi- and Low Volatiles	100% particulate	Not specified

Notes: (1) The 25% figure is arrived at by the Regulatory Guide 1.3, 1.4 assumption that 50% of the iodine inventory is released to containment, and half of this 50% plates out instantaneously.

TABLE 10. SUMMARY OF EVOLUTIONARY ALWR LICENSING SOURCE TERM UPDATE
(Continued)

	<u>Source Term Update</u>	<u>Existing Regulatory Source Term</u>
Aerosol Removal in PWR Containment from Sprays		
• 0-10 minutes after beginning of release	$\lambda = 100$ per hr.	$\lambda = 7.5$ per hr. ⁽²⁾
• 10-40 minutes after beginning of release	$\lambda = 50$ per hr.	$\lambda = 7.5$ per hr. ⁽²⁾
• 40-120 minutes after beginning of release	$\lambda = 5$ per hr.	$\lambda = 0.75$ per hr. ⁽²⁾
• 2-23 ⁽⁴⁾ hours after beginning of release	$\lambda = 1$ per hr.	$\lambda = 0.75$ per hr. ⁽²⁾
Mixing rate between sprayed and unsprayed volumes in PWR	$\lambda = 10$ unsprayed volumes per hour (i.e., 10 hr^{-1} as a removal coefficient) for first 2 hrs after beginning of release; $\lambda=2$ unsprayed volumes per hour thereafter	$\lambda = 2$ unsprayed volumes per hr (i.e., 2 hr^{-1} as a removal coefficient)
Suspended Aerosol Con- centration in BWR		
• 0-30 minutes after initiating event	40% of aerosol release to containment	10% of aerosol release to containment ⁽³⁾
• 30-120 minutes after initiating event	1% of aerosol release to containment ⁽⁵⁾	10% of aerosol release to containment ⁽³⁾

Notes:

- (2) Based on SRP 6.5.2 for aerosol removal.
- (3) Based on suppression pool scrubbing decontamination factor of 10 from SRP 6.5.5.
- (4) Since release begins at 60 minutes after accident initiation, 23 hours after the beginning of the release is 24 hours after accident initiation.
- (5) Based on a time integrated suppression pool decontamination factor of 100. Pool bypass needs to be considered to obtain overall decontamination.

TABLE 11. CALCULATION OF RELEASES TO CONTAINMENT ATMOSPHERE

	<u>Release From Fuel to RCS</u>	<u>Fraction of Fuel Release to RCS Which Escapes to Containment</u>	<u>Release to Containment</u>
Nobles	80%	1.0	80%
I, Cs	75%	0.3	22.5% ⁽¹⁾
Te	40%	0.3	12%
Ba, Sr, Sb, Ru	1%	0.3	.3%
Ce, La, Actinides	.01%	0.3	.003%

Notes: (1) It is recognized that the 22.5% figure may be more precise than warranted; necessary adjustment and/or round off can be made when the source term update is implemented.

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Appendix A

PHYSICAL PROCESSES ASSOCIATED WITH AEROSOL REMOVAL FROM THE CONTAINMENT ATMOSPHERE

The following is a brief discussion of the physical processes of aerosol mechanics which could be taken into account in establishing the source term. These processes provide a basis for crediting the "natural" depletion of fission product material in the containment atmosphere (analogous to the instantaneous 50 percent reduction of airborne iodine in containment assumed in TID 14844), as well as the depletion due to engineered safety feature operation in the calculation of fission product releases to the environment.

A.1 NATURAL DEPLETION

A.1.1 Agglomeration

Agglomeration is the process by which the size distribution of airborne particulate tends to shift with time to larger sizes until an equilibrium condition is reached. It is not a separate removal process, but affects several removal processes: sedimentation, pool scrubbing and spray removal. There are three agglomeration mechanisms that are generally treated, which include:

- | | | | |
|----|---------------|---|---|
| 1. | Brownian | - | the random movement of particles and the resultant collisions |
| 2. | Gravitational | - | the relative movement of particles of different size under the influence of gravity |
| 3. | Turbulent | - | the result of localized mixing with an effect of relative movement similar to gravitational |

In containment, Brownian agglomeration is important for submicron particles, while gravitational is important for particles larger than one micron. Turbulent agglomeration is generally unimportant in containment.

Multi-modal particle size distributions tend to become mono-disperse with time, with a distribution about a "self-preserving" particle size. The rate of growth up to that size becomes equal to the rate of sedimentation above that size. A typical, self-preserving particle size for containments (and incidentally, for ambient air) is about one μm .

A.1.2 Sedimentation

Sedimentation is deposition due to the effects of gravity on the particles, with accumulation generally on horizontal surfaces. In "stirred" systems, sedimentation still occurs, because if the system is closed, there is always a net downward movement of the particles. If the system is turbulent, both agglomeration and deposition will be enhanced.

The sedimentation removal constant, λ , is primarily a function of the aerosol concentration (or "cloud" density, m) and can be determined from analysis by mechanistic codes such as those mentioned in Section A.1.4 or through the use of correlations. Such a correlation was developed under IDCOR sponsorship and was further benchmarked by ARSAP.^{A-1} This correlation establishes functional relationships between a dimensionless removal rate constant $\Lambda = f(\gamma, x, \mu, h, \alpha, K_o, g, \rho)\lambda$ and a dimensionless aerosol mass density, $M = f(\gamma, \mu, h, \alpha, K_o, g, \rho)m$ where:

γ	=	collision shape factor
x	=	particle settling shape factor
μ	=	gas viscosity
h	=	effective settling height
α	=	density correction factor
K_o	=	normalized Brownian collision coefficient
g	=	gravitational acceleration
ρ	=	particle material density

Independent of the method chosen to quantify the sedimentation λ (either as constant or as a function of time), the value chosen should be a lower bound for the important sequence types and the plant design. A sedimentation λ of approximately 0.15/hr can be shown to be a reasonable (but still conservative) minimum using the correlation described above, and this is a good value to use as a conservative baseline.

The effects of hygroscopicity (discussed in the next section) would be to increase M , decrease ρ and cause α to approach unity. Hygroscopicity can be credited if it can be demonstrated that the containment atmosphere is maintained near saturation.

A.1.3 Hygroscopicity

Hygroscopicity is the term used to characterize the affinity of a substance for water. Substances that can maintain large quantities of water in solution are termed "hygroscopic." As described in Section 4.0 of the main report, the dominant chemical form of fission product cesium released to the containment in the course of a severe accident would be CsOH (i.e., cesium hydroxide), and CsOH is one of the most hygroscopic materials known. If in particulate form and in the aerosol size range it is exposed to atmospheres near saturation (saturation ratios greater than about 0.95), it can absorb factors of ten and even one hundred times its mass in water.^{A-2}

In containment, the effect of hygroscopicity is to increase the rate of particle growth and the sedimentation λ (see Section A.1.2) by typically a factor of two as long as the containment atmosphere is near saturation. This effect has been quantified both analytically and experimentally in the LACE series of experiments. Increasing the rate of particle growth would also be expected to increase the effectiveness of sprays as discussed in Section A.2.2.

A.1.4 Diffusiophoresis

As steam condenses on a surface, aerosol particles will migrate with the flux of water vapor moving to the surface and be deposited. This deposition process is referred to as diffusiophoresis. The importance of diffusiophoresis depends on the amount of condensation occurring in the accident sequence. If the surfaces in the containment are not cooled, the structures will tend to saturate thermally, steam condensation on the walls will slow, and the amount of diffusiophoretic deposition will decrease with time. Diffusiophoresis is a well-established phenomenon that is modeled in mechanistic computer codes of aerosol behavior such as CONTAIN^{A-3} and NAUA,^{A-4} as modified for incorporation into the NRC Source Term Code Package. Although not typically found to be the dominant deposition mechanism in severe accident analyses, diffusiophoresis can be an important contributor; it is not particularly sensitive to particle size, and, as a result, can be effective in the removal of an otherwise persistent airborne concentration of small aerosols even at fairly low condensation rates late in the accident.

The diffusiophoretic λ can be calculated by treating diffusiophoretic removal as if it were mathematically a "leak" from the unsprayed region. This mathematical model can be applied because the removal rate is not a function of the airborne particle size distribution. If the steam condensation rate in the unsprayed region is divided by the steam density, the result is a volumetric flow which can then be treated as a leak term, and as a diffusiophoretic λ when divided by the unsprayed region volume. This volumetric flow may be calculated in one of two ways, one with the steam density corresponding to the actual steam partial pressure (ideal estimate of diffusiophoretic deposition) and one with the density corresponding to the total pressure (a conservative estimate of diffusiophoretic deposition).

A.2 DEPLETION DUE TO ENGINEERED SAFETY FEATURES

A.2.1 Scrubbing in Suppression Pools

The attenuation of radionuclides in the pressure suppression pool is usually expressed as a "decontamination factor" (DF), which is defined as the ratio of the quantity injected into the pool divided by the quantity which escapes the surface of the pool. Although it has been generally accepted that large pools of water can be very effective in scrubbing contaminants from a gas stream passing through them (e.g. in iodine scrubbing in the spent fuel pool), it is recognized that the effectiveness varies significantly with a number of parameters. The Reactor Safety study (WASH-1400) assumed a DF of 100 for subcooled suppression pools, and 1.0 for steam saturated pools. Detailed models for the analysis of aerosol removal during gas transport through the suppression pool have been sponsored in recent years by the NRC (the SPARC code)^{A-5} and EPRI (the SUPRA code).^{A-6} A data base for code validation was developed with experimental programs at Battelle Columbus Laboratory sponsored by EPRI.^{A-7}

The results of analytical models, confirmed by experimental results, indicate that suppression pool scrubbing of aerosols depends on parameters associated with:

- The carrier fluid (steam/non-condensable gas ratio, temperature, mass flow rate)
- The entrained aerosol characteristics (size, material, density, solubility, aerodynamic characteristics)
- The injection configuration (submergence depth, orifice size and orientation, number of orifices in proximity),
- The water pool (subcooling, geometry, impurities)

Of these, the aerosol size is the most sensitive parameter. The observed DF, for example, varies over several orders of magnitude for aerosol sizes of interest in the region between 0.1 and 1.0 microns (see Figure A-1). A second important parameter shown in Figure A-1 is the condensable/non-condensable fraction of the carrier fluid. Large steam mass fractions result in large decontamination factors, while the minimum DF is calculated for dry

(hydrogen or air) gas flows. In contrast, the experiments showed the effect of pool saturation to be much less than anticipated, as a result of additional removal mechanisms (e.g., diffusiophoresis) associated with high steam fractions above the surface of the pool.^{A-8}

The injection configuration can have a significant effect on the pool entrance region. In contrast to the bubble rise region, where bubble dynamics and aerosol phenomena are well characterized by the models, the entrance region of the breakup of the gas stream entering the pool is more difficult to model. In the past, pool scrubbing models have either neglected the contribution of the entrance region to the overall pool decontamination factor, or accounted for it by using simplified approaches. However, pool scrubbing experiments^{A-9, A-10} have shown that scrubbing at the injection site can be significant and should be included in pool scrubbing analysis. An analytic model for aerosol scrubbing at pool injection sites^{A-11} developed under EPRI sponsorship concluded that scrubbing at the injection site can be appreciable. Though this analytic model requires validation, decontamination factors between 2 and 5, depending on noncondensable/condensable gas fraction, were calculated for ~0.3 μm particles. Battelle Northwest Laboratory has attempted to model the entrance effects, and has concluded that entrance effects would not extend beyond ten diameters of the vent pipe for horizontal vent injection configurations (i.e., Mark III-type suppression pools).^{A-12}

For any given set of these parameters, the existing models permit a reasonably accurate determination of the corresponding pool DF. With high steam content carrier gas, an anticipated aged particle size (e.g. 1-5 microns), and sufficient pool depth to minimize the effect of the entrance

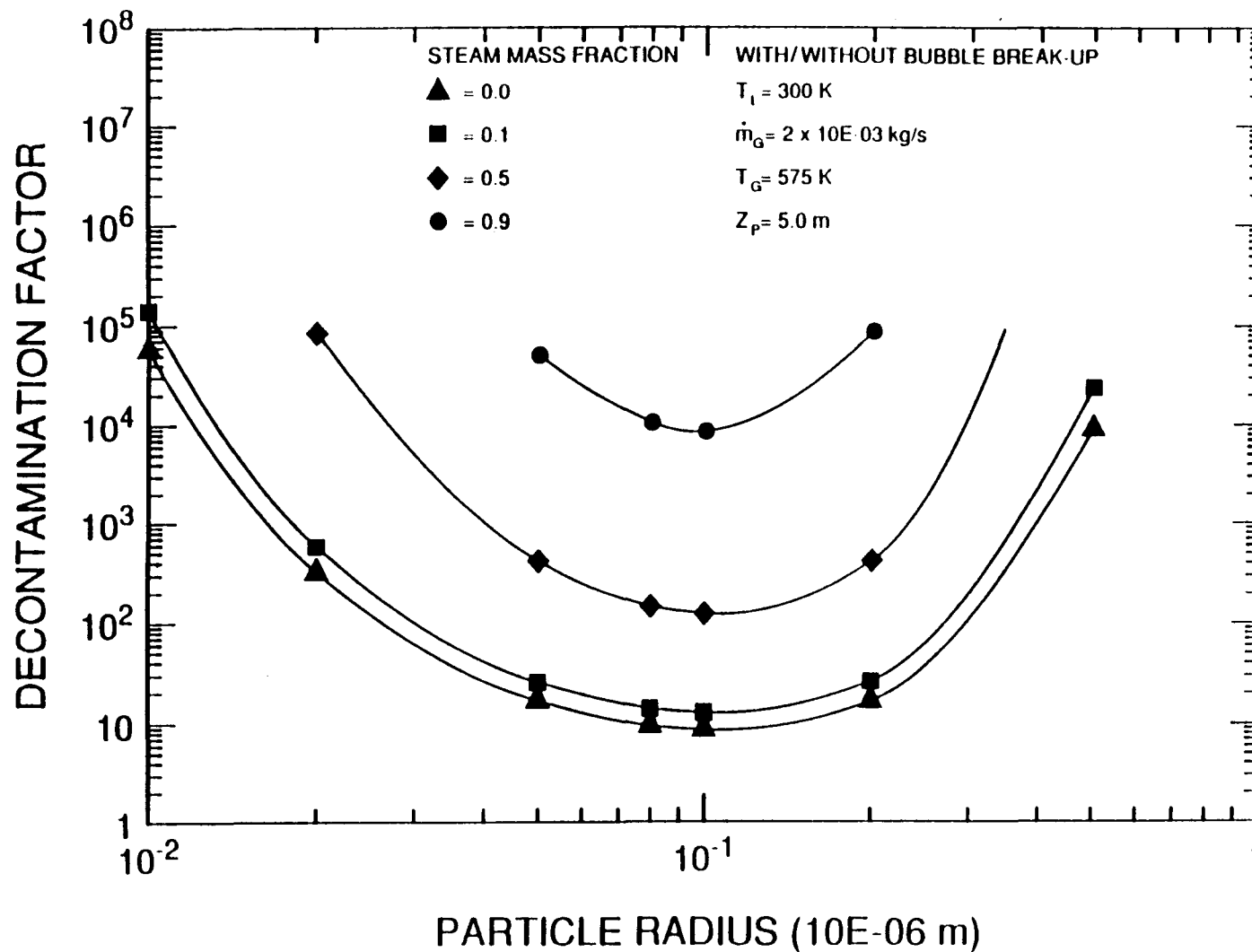


Figure A-1
EFFECT OF PARTICLE SIZE AND STEAM FRACTION ON DF
(Calculated by SUPRA Code)

region, pool decontamination factors well above 1000 are calculated, and have been observed experimentally.

During any specific accident sequence, however, several important variables may change significantly. In particular, fission products may be carried to the drywell and into the suppression pool in a hydrogen-rich gas mixture. Low steaming rates during such periods, however, would also reduce the total gas flow rate, resulting in a slower transport, and hence additional aging, of the fission product aerosols prior to injection into the suppression pool. The reduction in scrubbing efficiency resulting from the higher non-condensable gas fraction, therefore, is likely to be countermanded by an increase in efficiency resulting from increases in the aerosol size distribution.

Suppression pool models incorporated into integral severe accident codes will produce time varying suppression pool decontamination factors which quantitatively account for such changes in the important parameters. For the approach employed in this paper, i.e., bounding conditions encompassing a range of scenarios, a reduction in the overall DF to 100 for the suppression pool was used to account for high non-condensable gas fractions at times when fission product release rates are anticipated to be high.

With regard to the timing and rate of flow to the suppression pool as discussed in Section 6.3, the limiting accidents for containment leakage in the evolutionary BWR are those in which the release from the vessel is completely to the drywell. If the release from the RCS were accompanied by a large flow of steam and non-condensable gases, the fission product aerosols would be rapidly swept to the wetwell and subjected to decontamination in the suppression pool or would be carried to the walls of the drywell with condensing steam. However, for scenarios in which there is little or no water addition to the vessel during the period of core uncover, the quantity of gases and vapors released with the fission product aerosols may not be very large and limited sweepout or deposition can be expected during this period.

Because the evolutionary design does not have large pipes entering the vessel below the top of the core, the time required to result in core uncover in a pipe break accident is longer than in existing designs. Thus, the

earliest that fission product release from the fuel itself is expected to begin in an advanced BWR design is approximately one hour following accident initiation. Based on a review of calculations that have been made of the time period of core melting prior to extensive relocation to the lower plenum, one half hour is a reasonable bound on the period of fission product release that could occur for a sequence in which no water makeup is provided.

Following these periods of water boiloff and fission product release, additional core degradation is arrested either due to slumping of molten core debris into the lower plenum or due to water addition. The quantity of steam produced in removing the decay heat at one-hour following shutdown is 170,000 pounds per hour. At 30 psia this represents a turnover rate for the drywell of 10 volumes per hour. Thus the aerosol concentration in the drywell would decrease by a factor of "e" approximately every 6 minutes due to decay heat steaming alone. The steam from quenching of molten, slumping core debris would make the turnover rate even higher immediately after 30 minutes. For simplicity it is assumed that the sweepout following the period of fission product release is instantaneous.

During the 30 minute period of fission product release, some volumetric flowrate of gases must accompany the aerosols as they are transported to the drywell. In order to estimate the amount of sweepout of diffusiophoretic deposition that could occur during this period, the result of calculations with the Source Term Code Package of large pipe break accidents were examined. In these analyses, which were performed for an existing BWR, 20,000 pounds of water were boiled off during the period from zero percent core melting to approximately 80 percent core melting. Assuming a constant release rate of fission products and aerosols to the drywell over the one half hour time period results in an average airborne fraction of 40 percent of the total material released to the drywell.

If the form of the gas released to the drywell is steam, the removal mechanism in this period could either be sweepout or diffusiophoretic deposition. Recognizing that the concentration of aerosols in the drywell could be large in this time period, an estimate of the sedimentation removal rate was also made using the correlations developed for the MAAP code. However, when retention within the vessel is taken into account the estimated

removal rate for sedimentation is found to be an order of magnitude smaller than the sweepout term.

A.2.2 Spray Removal

A number of removal mechanisms can participate in the scrubbing of aerosols from the containment atmosphere by spray droplets:^{A-13}

- Inertial impaction occurs when the inertia of a particle is sufficient for it to cross the flow streamlines around the drop and thus contact the surface of the drop. Impaction is the dominant mechanism for large aerosol sizes (e.g., $> 10 \mu\text{m}$) but decreases very rapidly with decreasing size.
- Interception corrects inertial impaction predictions due to the finite size of the particle which allows collection even when the particle center of mass is on a trajectory which does not intersect the drop. Interception is dominant in the near submicron range.
- For very small aerosols, Brownian motion is the dominant mechanism. Unlike the efficiency of inertial impaction (which decreases rapidly with decreasing aerosol size) the collection efficiency of Brownian motion increases with decreasing size. Therefore, the overall collection efficiency of droplets passes through a minimum as a function of decreasing particle size. This minimum is typically in the near submicron range of particle size.

Diffusiophoresis and thermophoresis on the spray droplet are also mechanisms of particle collection but are not typically as important as the mechanisms discussed above.

The mechanisms for spray removal of aerosols have been incorporated in mechanistic containment codes such as the CONTAIN[3] code. Since spray droplet size also affects aerosol collection efficiency, it is necessary to characterize the size distribution of droplets in performing an analysis of spray removal; the distribution is then represented by an effective or

"characteristic" size. Particle size distributions are usually represented by perhaps ten or twenty "bins" (discrete size ranges), with the effect of the spray being calculated separately for a characteristic particle size for each bin. The same mechanisms that influence particle size growth for natural depletion (agglomeration and hygroscopicity, Sections A.1.1 and A.1.2, respectively) would also be expected to influence the effectiveness of spray, and are reflected in many current analytical models for spray removal.

In SRP 6.5.2, credit for spray removal of aerosols is permitted for PWRs but is limited to a conservative value of the removal coefficient as follows:

$$\lambda = \frac{3hFE}{2VD}$$

where h is the fall height of the spray drops, V is the containment building net free volume, F is the spray flow, and (E/D) is the ratio of a dimensionless collection efficiency E to the average spray drop diameter D . It is conservative to assume (E/D) to be 10 per meter initially (i.e., 1% efficiency for spray drops of one millimeter in diameter), changing abruptly to one per meter after the aerosol mass has been depleted by a factor of 50 (i.e., 98% of the suspended mass is ten times more readily removed than the remaining 2%).

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