# **SESSION 13**

# SOURCE TERMS AND ACCIDENT ANALYSIS

Wednesday: Co-Chairmen: July 27, 1994 R. R. Bellamy R. Zavadoski

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# **OPENING COMMENTS BY SESSION CO-CHAIRMAN BELLAMY**

In this plenary session we are going to discuss source terms and accident analysis. As you can see from the program, there are five papers in the session. Two of them are of international interest and two papers will discuss source term analyses. Another paper will get into a bit of a non-technical area. We will have a chance to hear some words this morning about the link between source terms and emergency planning. The author of the first paper is the principle author of a draft NUREG 1465. We are pleased to be able to continue our discussion of the Nuclear Regulatory Commission's revised accident source term.

#### REVISED ACCIDENT SOURCE TERMS FOR LIGHT-WATER REACTORS

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#### <u>Abstract</u>

This paper presents NRC revised accident source terms for light-water reactors (LWRs) which incorporate the significant severe accident research insights gained in this area over the last 15 years.

Current LWR reactor accident source terms used for licensing date from 1962 and are contained in Regulatory Guides 1.3 and 1.4. These specify that 100% of the core inventory of noble gases and 25% of the iodine fission products are assumed to be instantaneously available for release from the containment. The chemical form of the iodine fission products is also assumed to be predominantly elemental  $(I_2)$  iodine. These assumptions have strongly affected present nuclear air cleaning requirements by emphasizing rapid actuation of spray systems and filtration systems optimized to retain elemental iodine.

A proposed revision of reactor accident source terms and some implications for nuclear air cleaning requirements was presented at the 22nd DOE/NRC Nuclear Air Cleaning Conference. A draft report (NUREG-1465) was issued by the NRC for comment in July 1992. Extensive comments were received. While most have been favorable, the most significant comments involve (a) release fractions for both volatile and non-volatile species in the early in-vessel release phase, (b) gap release fractions of the noble gases, iodine and cesium, and (c) the timing and duration for the release phases. The final source term report is currently in preparation and is expected to be issued in late 1994. Although the revised source terms are intended primarily for future plants, current nuclear power plants may voluntarily request use of revised accident source term insights as well in licensing.

This paper emphasizes additional information obtained since the 22nd Conference, including contractor studies on fission product removal mechanisms, results obtained from improved severe accident code calculations and resolution of major comments, and their impact upon the revised accident source terms.

Revised accident source terms for both BWRS and PWRS in terms of fission product composition, magnitude, timing and iodine chemical form are presented. Related regulatory activities in regard to severe accident source terms are also discussed.

# I. Introduction and Background

Radionuclide releases, that is, the type, quantity, timing and energy characteristics of the release of radioactive material from reactor accidents ("source terms") are deeply embedded in the regulatory policy and practices of the U.S. Nuclear Regulatory Commission (NRC). The NRC's reactor site criteria<sup>(1)</sup> require for licensing purposes that an accidental fission product release from the core into the containment be postulated to occur and that its radiological consequences be evaluated assuming that the containment remains intact but leaks at its maximum allowable leak rate.

The characteristics of the "source term" into the containment, which must be distinguished from a release to the environment, is contained in Regulatory Guides 1.3 and  $1.4^{(2,3)}$ , but is derived from the 1962 report TID-14844<sup>(4)</sup>, and consists of 100% of the core inventory of noble gases and 50% of the iodines (half of which are assumed to deposit on interior surfaces very rapidly).

Severe accident research insights<sup>(5)</sup> have confirmed that although the TID-14844 release is very substantial and has resulted in a very high level of plant capability, nonetheless, based upon the large amount of information obtained on severe accidents since the publication of TID-14844 more than thirty years ago, the present recipe is no longer compatible with a realistic understanding of severe accidents. It has both conservative and non-conservative aspects. For example, the assumption of an instantaneous appearance of a large fraction of the core inventory of noble gas and iodine isotopes within containment has long been recognized as highly conservative, whereas neglect of other important nuclides released into containment, such as cesium, may be non-conservative.

A major NRC research effort began about 1981 and has been under way since then to obtain a better understanding of fission product transport and release mechanisms in light water reactors (LWR) under severe accident conditions. This improved methodology on severe accident source terms has been reflected in NUREG-1150<sup>(6)</sup> which provides an updated risk assessment for five U.S. nuclear power plants. An assessment of the chemical form of iodine found within containment as a result of a severe accident has also been carried out<sup>(7)</sup>.

The NRC has issued a draft report for comment<sup>(8)</sup> proposing revised LWR reactor accident source terms. These proposed accident source terms, including a discussion of some implications for nuclear air cleaning requirements, was presented at the 22nd DOE/NRC Nuclear Air Cleaning and Treatment Conference.<sup>(9)</sup>

#### II. <u>Revised Accident Source Terms</u>

The timing, magnitude in terms of fractions of the core inventory released into containment, and the composition of the proposed revision of accident source terms were based upon a range of severe accident sequences studied in NUREG-1150 and included release fractions for a complete core-melt accident, including core-concrete interactions and

releases associated with late in-vessel volatilization. A revised iodine chemical form<sup>(7)</sup> was also incorporated. The draft report also included a quantitative discussion of non fission product aerosols released into the containment from reactor vessel materials and coreconcrete interactions. It must be emphasized that the formulation of the revised source term was intended to represent the generalized progression of a severe accident in order to assure that a plant design is capable of adequately mitigating such accidents. The characteristics of the revised source term were not intended to be an accurate representation of any single accident sequence, but rather combines aspects of a number of core-melt sequences.

Comments on the draft revised source term were solicited from an internationally recognized group of experts in severe accident source terms, as well as from the nuclear industry and members of the public. Over 200 separate comments were received from more than 20 individuals and organizations. The comments were generally favorable and expressed the view that the revised source term incorporated severe accident research insights and represented a major advance over the older source term employed in TID-14844. However, a number of comments expressed the view that the early in-vessel release fractions might be low, whereas the ex-vessel release fractions were believed too high. Commenters also stated that the release fractions for the low volatile nuclides, both in-vessel as well as ex-vessel, were believed to be too high. Release fractions for the gap release phase were also considered high. Comments were also received to the effect that iodine chemistry was rather complex, and that organic iodine was not addressed in the draft report. Finally, several commenters stated that the report should include a better treatment of removal mechanisms, and that showing releases for a dry reactor cavity was inappropriate for a number of advanced reactor designs that were proposing to maintain a flooded reactor cavity.

As part of the effort to finalize the source term, the NRC also sponsored contractor studies to develop more realistic models of predicting removal of fission products within containment by natural removal mechanisms as well as by engineered features such as sprays and suppression pools. At the present time, studies providing improved models of aerosol scrubbing by a water pool overlying core debris<sup>(10)</sup> and aerosol removal by containment sprays<sup>(11)</sup>, have been completed and issued. Additional studies on aerosol removal by BWR suppression pools and aerosol removal in containment by natural removal processes are still in progress.

In addition, studies using the  $MELCOR^{(12)}$  code have been performed for several sequences at the Peach Bottom<sup>(13)</sup> and Surry<sup>(14)</sup> nuclear power plants. While results from these studies showed general agreement with those of draft NUREG-1465, the duration of the gap release phase for BWR reactors was reduced from 1.0 to 0.5 hours, based on these studies<sup>(13)</sup>.

The final version of NUREG-1465 is currently being readied for publication. Revisions have been incorporated as a result of the comments received as well as the additional studies noted above. The major changes in the final source term report compared with the draft version are summarized below:

- (1)Release fractions for the non-volatile fission products during the early in-vessel phase, particularly cerium and lanthanum, were significantly reduced. Release fractions for the same nuclides during the ex-vessel phase were also reduced somewhat. The release fractions used in the draft source term report were based on mean values of the NUREG-1150 distributions to estimate release fractions for all nuclides. Since release estimates for the nonvolatile nuclides showed a very large degree of uncertainty (spanning over four orders of magnitude) compared to the volatile nuclides, the mean value was unduly influenced by the extreme upper end of the range, and was in excess of other measures of the distribution, such as the 75th percentile. Recent research results<sup>(15)</sup> since the issuance of NUREG-1150 also indicate that of the non-volatile fission products has release been overestimated. For the final source term report, the 75th percentile of the distribution was selected to estimate the release fractions for the non-volatile nuclides, since it bounded the bulk of the data, without being unduly conservative.
- (2) The duration of the BWR gap release phase was reduced from 1.0 to 0.5 hours, as noted above.
- A discussion of organic iodide formation, not addressed in the (3)draft report, was incorporated in the final version. Organic iodine, while representing only a small fraction of the iodine released, can be important in the evaluation of the radiological consequences of design basis accidents because it is not readily removed by sprays or filters. It appears primarily as CH<sub>3</sub>I, and is produced over time primarily as a result of the interaction of elemental iodine with organic compounds present in containment. An analysis and review of containment experiments<sup>(16)</sup> indicated that about 3.2 percent of the airborne iodine would be converted to organic iodides within the first two hours. The final source term report has adopted a conversion of 4 percent of the airborne iodine to organic form. Where the pH is controlled to a value of 7 or greater, elemental iodine would be no greater than about 5 percent of the total released. Hence, where pH control is maintained, organic iodide would represent about 0.20 percent of the total iodine released.
- (4) A revised grouping of the nuclides, shown in Table 1, was adopted based on grouping barium and strontium together, because of similarity, rather than separately, as in the draft report.
- (5) Quantitative estimates of fission product removal mechanisms, will be deleted from the final report. Since fission product removal is highly plant specific, treatment of this important area should be done on a case specific basis.
- (6) Numerical tabulations of non fission product aerosols masses released into containment will be deleted from the final report, based on studies<sup>(13)</sup> indicating that estimates in the draft report may have been significantly overestimated. Since reliable data may not be presently available, non fission product aerosols will be addressed in the final report qualitatively.

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TITLE	ELEMENTS in GROUP						
Noble Gases	Xe, Kr						
Halogens	I, Br						
Alkali Metals	Cs, Rb						
Tellurium group	Te, Sb, Se						
Barium, Strontium	Ba, Sr						
Noble Metals	Ru, Rh, Pd, Mo, Tc, Co						
Lanthanides	La, Zr, Nd, Eu, Nb, Pm, Pr, Sm, Y, Cm, Am						
Cerium group	l Ce, Pu, Np						

Table 1 Revised Radionuclide Groups

Revised accident source terms, including composition, magnitude, ti ing and duration, are shown in Tables 2 and 3 for BWRs and PWRs, respectively.

	Gap Release	Early In-Vessel	Ex-Vessel	Late In-Vessel
Duration (Hours)	0.5	1.5	3.0	10.0
Noble Gases	0.05	0.95	0	0
Halogens	0.05	0.25	0.30	0.1
Alkali Metals	0.05	0.20	0.35	0.1
Tellurium	0	0.05	0.25	0.005
Barium, Strontium	0	0.02	0.1	0
Noble metals	0	0.0025	0.0025	0
Cerium group	0	0.0005	0.005	0
Lanthanides	0	0.0002	0.005	0

Table 2 Revised BWR Releases Into Containment\*

\* Values shown are fractions of core inventory.

The rate of release of fission products into the containment is assumed to be constant during the duration time of each phase. The releases shown for the ex-vessel phase are assumed to be for a dry reactor cavity having no water overlying any core debris. Where water is covering the core debris, aerosol scrubbing will take place and reduce the quantity of aerosols entering the containment atmosphere.

	Gap Release	Early In-Vessel	Ex-Vessel	Late In-Vessel
Duration (Hours)	0.5	1.3	2.0	10.0
Noble Gases	0.05	0.95	0	0
Halogens	0.05	0.35	0.25	0.1
Alkali Metals	0.05	0.25	0.35	0.1
Tellurium	0	0.05	0.25	0.005
Barium, Strontium	0	0.02	0.1	0
Noble metals	0	0.0025	0.0025	0
Cerium group	0	0.0005	0.005	0
Lanthanides	0	0.0002	0.005	0

Table 3 Revised PWR Releases Into Containment\*

\* Values shown are fractions of core inventory.

# III. <u>Regulatory Applications</u>

When issued final, the revised source terms in NUREG-1465 are intended to be applied for future plants and will not be backfit for presently licensed plants. One advanced design (Combustion Engineering System 80+) has used the source terms in the draft NUREG-1465 for licensing purposes, and the NRC staff has approved, subject to revision when the final report is issued.

The NRC staff has also proposed<sup>(17)</sup> that for evaluation of design basis accidents (DBA) for the evolutionary and passive light-water reactor designs, only the releases associated with the gap and early in-vessel release phases will be used. The NRC staff has indicated that it considers the inclusion of the ex-vessel and late in-vessel releases to be unduly conservative for DBA purposes. Such releases would only result from core damage accidents with vessel failure and core-concrete interactions. For evolutionary and passive light-water reactors, the estimated frequencies of such scenarios are low enough that they need not be considered credible for the purpose of meeting 10 CFR Part 100.

However, the staff has also noted that equipment and features needed for severe accident prevention, mitigation, and sampling should be designed to provide a reasonable level of confidence that they will operate in a severe accident environment. Such an environment would include the ex-vessel and late in-vessel releases, in addition to those for DBAs.

After final issuance, licensees of current plants may voluntarily request use of the revised source terms in NUREG-1465 for plant applications. Several licensees have expressed interest in such usage, and the NRC staff is preparing a plan to consider such applications.

# IV. Conclusions

Revised reactor accident source terms, when issued in final form, will reflect an improved understanding of fission product releases into containment for a spectrum of degraded core and severe accidents having regulatory application to light-water reactors.

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#### DISCUSSION

- **MISHIMA:** Do you think these source terms are applicable to the effects of explosions on spent fuel that would severely fragment the spent fuel, therefore increasing the surface area allowing release?
- **SOFFER:** That was not what we had in mind. The source terms in NUREG 1465 were developed from reactor accident sequences involving core heatup and fuel melting. It is unlikely, in my opinion, to be applicable for spent fuel explosions. For one thing, spent fuel does not have much in the way of noble gases or much in the way of iodines, which are, of course, the things that have traditionally been of greatest concern in reactor accidents. For another, in fragmenting and exploding fuel, you are talking about releasing aerosolized quantities of fuel particles, including uranium, transuranides, et cetera, which are not the kinds of things that one would get in large quantities in the normal heatup of fuel. So my reaction is, probably not.
- MISHIMA: What about the volatile materials that are present after five year cooling?
- **SOFFER:** To the extent that there is cesium there, it may be okay for that. But to the extent that you are going to get plutonium and neptunium, etc., I suspect that that is not the ideal way of doing it.
- **FIRST**: You made a point about accepting applications from plants to use the new source

terms. What are the advantages of plants electing the new source term?

- **SOFFER:** Based on the limited information we have from utilities, the industry appears to believe that there are a number of advantages in the timing aspects; that is, in getting away from an instantaneous release. For example, at the present time, secondary containments in a BWR have to turn on emergency ventilation systems immediately, and are required to get down to a negative pressure in order to receive credit for that function to be accepted as a secondary containment. This puts a rather high load on the diesels, and puts a large strain on the ventilation systems. There are people who believe this is not warranted in view of timing considerations. There are a number of people who feel the timing aspects of the source term provide advantages with regard to improved reliability, valve closure time, easier loading on diesels, things of this kind. There may be other applications as well.
- HULL: As a Long Islander and someone who has been inside the business, I realize you are probably not the person to answer this question, but I will ask it anyway. Shoreham is gone, so it is too late for that, but do you see any of this leading to a relaxation of the ten mile EPZ, which effectively did in Shoreham (if any one requirement did)?
- **SOFFER:** That is going to be the subject, I believe, of another paper in this session, and I would like to hear what the other author will say. Emergency planning has been based on a spectrum of accidents that considered not only the design basis accident, but considered very severe accidents where containments are likely to fail. The revised source terms were developed for plant design purposes where the containment is assumed not to fail to incorporate a better understanding of degraded core accidents. These source terms were not intended to be used for emergency planning.
- **KOVACH; L.:** Is there to be any revision of fuel handling accidents, in addition to vessel accidents, in the new source term?
- **SOFFER:** The fuel handling accident is not specifically covered in the revised source term; however, revised gap release fractions are provided. This might provide a basis for reconsidering the fuel handling accident. There is going to be a new revision of gap activity releases. And this could lead the way for a revision of fuel handling accidents.

KOVACH, L.: That would be another document?

**SOFFER**: That is right.

#### POTENTIAL RADIONUCLIDE EMISSIONS FROM STACKS ON THE HANFORD SITE, PART 1: DOSE ASSESSMENT

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#### Abstract

On February 3, 1993, the U.S. Department of Energy, Richland Operations Office (RL) received a Compliance Order and Information Request from the Director of the Air and Toxics Division of the U.S. Environmental Protection Agency (EPA), Region 10. The Compliance of the requires RL to evaluate all radionuclide emission points at the Hanford Site to determine which are subject to continuous emission monitoring requirements in Title 40, <u>Code of Federal Regulations</u>, Part 61, (40 CFR 61) Subpart H, and to continuously monitor radionuclide emissions in accordance with requirements in 40 CFR 61.93. The Information Request required RL to provide a written Compliance Plan to meet the requirements of the Compliance Order. A Compliance Plan was submitted to EPA, Region 10, on April 30, 1993.

The Compliance Plan specified that a dose assessment would be performed for 84 Westinghouse Hanford Company (WHC) stacks registered with the Washington State Department of Health (WAC 246-247) on the Hanford Site. Stacks that have the potential emissions to cause an effective dose equivalent (EDE) to a maximum exposed individual (MEI) greater than 0.1 mrem  $y^{-1}$  must be monitored continuously for radionuclide emissions. Five methods were approved by EPA, Region 10 for performing the assessments: Release Fractions from Appendix D of 40 CFR 61, Back Calculations Using A HEPA Filtration Factor, Nondestructive Assay of HEPA Filters, A Spill Release Fraction, and Upstream of HEPA Filter Air Concentrations. The first two methods were extremely conservative for estimating releases. The third method, which used a state-of-the-art portable gamma spectrometer, yielded surprising results from the distribution of radionuclides on the HEPA filters. All five methods are described.

Assessments using a HEPA Filtration Factor for back calculations identified 32 stacks that would have emissions that would cause an EDE to the MEI greater than 0.1 mrem  $y^{-1}$ . The number was reduced to 15 stacks when the other methods were applied. The paper discusses reasons for the overestimates.

#### I. Introduction

On December 15, 1989, EPA promulgated in the <u>Code of Federal</u> <u>Regulations</u> (CFR) the National Emission Standards for Emissions of Radionuclides Other Than Radon From Department of Energy Facilities--40 CFR 61, Subpart H<sup>(1)</sup>. Subpart H requires the emissions of radionuclides from the U.S. Department of Energy (DOE) facilities shall not exceed those amounts that would cause any member of the public to receive an effective dose equivalent (EDE) of 10 mrem in a year. In addition, potential emissions of radionuclides from a facility that could cause an EDE to a maximum exposed individual to exceed 0.1 mrem y<sup>-1</sup> require continuous monitoring.

To determine compliance, section 61.93 provides requirements for monitoring of radionuclide emissions from point sources. These monitoring requirements became effective upon promulgation of the regulation on December 15, 1989. Also, DOE Richland Operations Office (RL) as defined in 40 CFR 61 is an "owner or operator" of a "facility," i.e., the Hanford Site (see Figure 1). On February 3, 1993, RL received a Compliance Order and Information Request from the Director of the Air and Toxics Division of the U.S. Environmental Protection Agency (EPA), Region 10. The Compliance Order required RL to evaluate all radionuclide emission points at the Hanford Site to determine which are subject to continuous emission measurement requirements in 40 CFR 61, Subpart H, and to continuously measure radionuclide emissions in accordance with 40 CFR 61.93. The Information Request required RL to provide a written Compliance Plan to meet the requirements of the Compliance Order. A Compliance Plan was submitted to EPA, Region 10, on April 30, 1993. The Compliance Plan set as one ' Complete assessment of the 84 Hanford Site of the milestones: registered stacks under management by WHC by December 17, 1993.



Figure 1 Hanford site.

The purpose of this document is to present the dose assessment results for the registered stacks on the Hanford Site for potential emissions (i.e., emissions with no control devices in place). Further, the document will identify those stacks requiring continuous monitoring (i.e., the EDE from potential emissions >0.1 mrem  $y^{-1}$ ).

#### II. Scope

The stack assessment of potential emissions was performed on 84 registered stacks on the Hanford Site. These emission sources represent individual point sources presently registered under Washington Administrative Code 246-247 with the <u>Washington Department of Health</u>. Unfortunately, for most stacks potential emissions are not sampled. For these stacks, other methods were developed and submitted to EPA, Region 10 for approval. The methods used in assessing the potential emissions from the stacks are described below.

#### III. Methods for Calculating Potential Stack Emissions

The calculation of potential emissions for the registered stacks were made using five methods approved for use by the EPA, Region 10:

- 1. Source Term Estimation Based on release factors in Appendix D
- 2. Back Calculations Based on a Decontamination Factor of 3000<sup>n</sup>, where n equals the number of HEPA filter banks in series
- 3. Nondestructive Assessment (NDA)
- 4. Upstream of HEPA Filter Air Concentration Measurements
- 5. Spill Release Fraction (296-Z-5)

Calculations based on the first two methods are considered extremely conservative. The last three methods should provide a realistic estimate of the actual potential releases. When the assessment of stack emissions based on Appendix D or from back calculations could indicate that the potential emissions would result in an EDE exceeding 0.1 mrem  $y^{-1}$ , while one of the last methods would result in an EDE less than 0.1 mrem  $y^{-1}$ , the more realistic method results would be accepted over the first two conservative methods.

#### 1. Appendix D Release Factors

The potential emissions for a system without control devices can be estimated based on factors presented in 40 CFR 61 Appendix D: for gases the release factor is 1, for liquids or particulate solids the release factor is  $1 \times 10^{-3}$ , and for solids the release factor is  $1 \times 10^{-6}$ . This method is extremely conservative, because accidents involving liquids and loose particulates have a release fraction orders of magnitude less than the  $1 \times 10^{-3}$  release fraction. In an accident condition, a spill of powder from a 1 m height of the entire inventory would release under extremely conservative conditions  $4 \times 10^{-5}$  release fraction and a liquid spill from the same would release approximately  $1 \times 10^{-6}$  release fraction. A  $1 \times 10^{-3}$  release fraction is used for powders and liquids. In cases where the assessment was performed on waste tanks, if the inventory was covered by supernate, the supernate was evaluated using the release factor for liquids while the covered salt cake was estimated using a release factor for solids.

#### 2. Back Calculations Based on a Decontamination Factor of 3000<sup>n</sup>

This method for estimating potential emissions assumes the Nuclear Air Cleaning Handbook<sup>(2)</sup> decontamination factor (DF) of 3000 (DF<sup>-1</sup>/(1-0.9997)) for 0.3 $\mu$  by a HEPA filter. The method assumes that each bank in series acts independently of a preceding bank and will remove contamination with the same 3000 decontamination factor. system with n banks of HEPA filters For а in series, the decontamination factor is 3000<sup>n</sup>. This method can be conservative for When processing no longer occurs, the a contaminated system. resuspension of contamination downstream of the HEPA filters can dominate the airborne releases from a facility. Multiplying these releases by 3000<sup>n</sup> will overestimate the potential emissions by orders of magnitude.

#### 3. Nondestructive Assessment (NDA)

In September 1992, EPA, Region 10, concurred with a WHC proposal for a test method to measure radioactive particulate emissions (particularly gamma emitters from high-efficiency particulate air [HEPA] filters) in stack exhauster systems across the Hanford Site (see Part II, Barnett, 1994<sup>(3)</sup>). A nondestructive assessment (i.e., in situ) method was developed, tested, and implemented using portable lowresolution (NaI) gamma spectroscopy instrumentation<sup>(4)</sup> to evaluate gamma emissions from operating HEPA filters. Guidance for the developed method comes from the 40 CFR 61, Subpart H and Appendix B, Method 114<sup>(5)</sup>.

# 4. Upstream of HEPA Filter Air Concentration Measurements

The upstream air concentrations provide direct information on the potential emissions from a facility. This method can be applied by using Continuous Air Monitoring (CAM) data, inserting air sample probes for upstream measurements, or radiological analysis of removed HEPA filters. CAM data can be used if the data is taken from the process area that contains the radionuclide inventory. Air measurements may also be collected in the ventilation system upstream of the HEPA filters. In this case, a sampling port is selected, a probe inserted, and air concentrations are measured. The final method is the sampling of the furthest upstream HEPA filters for the facility. In this case, one or more HEPA filters are removed and the radiological content is analyzed either by destructive assay or by a gamma spectrometer. The airflow during the time the HEPA filter is online is used to determine the annual release rate. These methods are technically based in that the measurements represent the potential concentrations emitted from a facility without control devices.

#### 5. Spill Release Fraction (296-Z-5)

In the facility venting to the 296-Z-5 stack, the only potential emissions occur when a canister of PuO<sub>2</sub> powder is repackaged. No emissions are expected from contamination in the ventilation system for the facility because it is essentially uncontaminated. When a canister is repackaged, the double canisters containing 1,500 g of PuO<sub>2</sub> are opened in a hood and the powder is poured into another canister. The canister is sealed and put into another canister and it is sealed. The

pouring could cause a release of  $PuO_2$  to the air. To estimate the potential maximum release, observed spill release fractions for depleted uranium oxide spills (Sutter et al. 1981<sup>(6)</sup>) was used. A maximum release fraction value of 4.0 x 10<sup>-5</sup> was used. This number is extremely conservative because it represents a release fraction for a 1 m release height for the spill. The actual release height is a maximum of 0.15 m. The spill release fraction was only used for the 296-Z-5 stack.

#### IV. CAP88 Dose Modeling

A potential source term was calculated for the 84 registered stacks using one of the five approved methods. Once the potential source terms were determined, the source term, location, and stack height were used with Hanford Site meteorology in the CAP88 computer model (Beres 1991<sup>(7)</sup>) to predict the EDE for the maximum exposed individual.

The CAP88 model is able to incorporate plume rise from the stack based on the flow rate and stack diameter. After leaving the stack, the plume is modeled to disperses based on meteorological conditions. Modeled concentrations are decreased based on dry deposition (dry deposition velocity = 1.8 cm/s.) However, wet deposition is ignored because of the low incident of precipitation on the Hanford Site. The modeled concentrations are used to calculate an EDE for an offsite MEI by summing the product of the EDE for each isotope with its activity.

$$\dot{H}_{total} = \sum (EDE_{(i)} \times A_{(i)}) = mrem y^{-1}$$
 (1)

where:

Normalized source terms were used in CAP88 for each of the major areas on site (i.e., 100, 200, 300, 400, and 600 Areas). The 5-year average of Hanford Site meteorological data (1983-1987) taken on the site was used to compute normalized air concentrations, i.e, X/Q, where X is the air concentration (curies/m<sup>3</sup>) normalized by the source term Q (curies/s). Two stack release heights were used: 89 m (200 ft stack with a 100 ft plume rise) and 10 m (33 ft). The following input was reeded to model an individual stack: the potential radiological source terms, the stack release height was chosen, and the location of the stack in one of the major areas. Potential dose data was produced by the CAP88 model for each registered stack.

#### V. Results of Dose Assessment

Of the 82 registered stacks assessed, 41 were assessed using release fractions from Appendix D, 31 using back calculations, nine stacks using NDA, two using upstream air samples, and one using the powder release fraction. Fifteen stacks (see Table 1) were identified to have potential emissions that could cause an effective

Stack Number	Facility	Number of HEPA Filters
291-A-1 <sup>b</sup>	PUREX	3
296-A-1	PUREX	3
291-B-1 <sup>b</sup>	B Plant	2
291-Z-12	PFP	1-3°
296-Z-3	PFP	2
296-A-22 <sup>b</sup>	Tank Farms	2
296-A-27	Tank Farms	2
296-A-29	Tank Farms	2
296-A-40	Tank Farms	2
296-P-16	Tank Farms	2
296-A-22 <sup>b</sup>	Tank Farms	2
296-P-23	Tank Farms	2
296-S-7W	ERO	2
296-S-15	Tank Farms	2
308-GL-EX	308 Building	3
340-NT-EX <sup>b</sup>	340 Building	2

Table 1 Designated stacks.

<sup>a</sup>Stacks with potential emission to cause a dose >0.1 mrem y<sup>-1</sup>.

<sup>b</sup>Original seven designated stacks.

<sup>c</sup>Stages of filtration depends on exhaust pathway.

dose equivalent >0.1 mrem  $y^{-1}$ . One of the original seven designated stacks, 291-T-1, was assessed by NDA and reduced to a nondesignated stack<sup>(8)</sup>.

To evaluate the five different methods, each method was compared to the results of back calculations for all stacks. The back calculations were performed for all stacks and the results indicated emissions from 32 stacks would have caused an EDE that would have exceeded 0.1 mrem  $y^{-1}$  and required the stacks to provide continuous When a stack reported a non detectable release, the monitoring. minimum detection limit was used as the release for the stack. Α comparison made for a subset of 30 stacks using release factors from Appendix D with back calculations showed no correlation between the two methods (Figure 2). Further, 21 of the 30 back calculations potential emission exceeded the release fractions emissions. In only four comparisons did the release fractions from Appendix D identify a designated stack when the back calculation indicated a nondesignated.



Figure 2 Doses from back calculations compared to release fractions from Appendix D (\* - Release Fraction Appendix D, • - Back calculations).

The comparison between NDA and back calculations showed that back calculations overestimated the potential releases for all nine stacks by at least four orders of magnitude (Table 2 and Figure 3). (Four of the stacks [see Table 3] reported nondetection of a release. For these stacks the minimum detection limit was used for back calculations.) If back calculations had been used instead of NDA, all nine stacks would have required continuous monitoring. Upstream air sample method for two of the stacks showed that the back calculations overestimated the emissions by three orders of magnitude. For the single stack that used the powder release fraction, the back calculations overestimated the release by four orders of magnitude.

Stack Number	NDA (mrem y <sup>-1</sup> )	Back Calculation (mrem y <sup>-1</sup> )	LOG <sub>10</sub> (Difference (mrem y <sup>-1</sup> ))
291-T-1	4E-3	4E+2	5
296-T-13	3E-4	3E+1	5
296-B-13	5E-8	2E+0 <sup>a</sup>	8
296-G-1	3E-8	3E-1ª	7
296-S-2	1E-5	1.2E-1	4
296-S-16	2E-6	1.0E-1	5
296-W-3	2E-9	3E+0 <sup>a</sup>	9
308-ET-EX	3E-5	1E+2	7
437-MN&ST	2E-3	2E+1 <sup>a</sup>	4

Table 2 NDA Comparison with back calculations.

<sup>a</sup>Minimum detection limit used for back calculations.



Figure 3 Doses from NDA compared to back calculations.

Stack Number	Method	Number of HEPA	Nuclide	Unabated (Ci)	Release (Ci)	DF
291-T-1	NDA	Prefilter, 2 HEPAs	Sr90	1.6E-3	1.2E-5	190
			Cs137	1.7E-3	1.3E-5	130
		1	Pu239,240	2.0E-3	2.2E-5	110
			Am241	2.0E-4	2.0E-6	100
296-T-13	NDA	2	Pu239,240	1.7E-4	2.0E-6	90
<b>296-</b> T- <b>1</b> 1	Upstream	2	total beta	1.5E-5	4.3E-7	30
296-5-2	NDA	2	total alpha	2.1E-6	8.4E-9	240
			total beta	3.5E-6	1.4E-8	250
296-S-7W	Upstream	2	Pu239,240	3.2E-5	7.5E-7	40
			Am241	3.9E-4	4.2E-6	90
296-5-16	NDA	1	total alpha	3.1E-7	1.7E-9	185
			total beta	9.8E-6	5.3E-8	180
308-ET-EX	NDA	2	total alpha	9.3E-7	9.2E-8	10
ann an			total beta	1.8E-7	3.4E-7	0.5
437-MN&ST	NDA	2	total alpha	2.0E-5	1.9E-7	105
			total beta	7.8E-5	7.4E-7	106

Table 3 Apparent decontamination factors for nondesignated stacks.

When upstream measurements or NDA were performed, an apparent decontamination factor was computed for eight nondesignated stacks having releases above the detection limit. The term apparent has been

used because an actual decontamination factor could not be computed. An actual decontamination factor would have to be calculated for a variety of particle sizes with air concentration measurements before and after the filters. The apparent decontamination factor was formed using either the estimated annual air concentration or the loading on the filters ratioed with the measured releases. The measured releases were <2E-5 curies per year for a radioisotope for all the stacks. The unabated emissions for these stacks were less than 3E-3 curies  $y^{-1}$ . For one stack 308-ET-EX a DF of 0.5 was computed (i.e., more released than was assessed on the HEPA filters). For some of the designated stacks the DFs, if measured, would produce larger DFs than noted in Table 3.

#### VI. Conclusions

By using the five approved methods approved by EPA, Region 10 instead of only the original back calculation method for assessing the 84 WHC stacks, the number of stacks requiring continuous monitoring was reduced from 32 to 15.

The intercomparison between results showed that no correlation existed between back calculations and release fractions from Appendix D results. Also, the NDA, upstream air samples, and powder release fraction method results were at least three orders of magnitude lower than the back calculations results.

The most surprising results of the assessment came from NDA. NDA was found to be an easy method for assessing potential emissions. For the nine stacks assessed by NDA, all nine would have required continuous monitoring when assessed by back calculations. However, when NDA was applied, all stacks had potential emissions that would cause an effective dose equivalent below the >0.1 mrem y<sup>-1</sup> standard.

Apparent decontamination factors were calculated for eight nondesignated stack emissions above the detection limit. These apparent decontamination factors ranged from 0.5 to 250.

#### Acknowledgments

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<sup>6</sup>Sutter, S. L., Johnson, J. W., and Mishima, J., 1981 "Aerosols generated by free fall of powders and solutions in static air," NUREG/CR-2139, PNL-3786 RH.

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# DISCUSSION

- KOVACH, L.: The in-place permitted leak rate for a HEPA filter bank is 0.05% maximum. The efficiency of the filter itself is a minimum of 99.97%. Thus, permissible in-place efficiency is always lower than 99.97%.
- DAVIS: No comment.
- **KOVACH, L.:** Presence of contamination downstream of HEPA banks is always an indication of a bypass of HEPA filter banks (possibly during change out or before installation).
- **DAVIS:** The presence of contamination downstream of the HEPA filters comes from: (1) older facilities (from the early 40's) operated with no filtration, causing contamination of the ducting, (2) for processing facilities, operations have now ceased but contamination was caused during the processing period, (3) during change-out some contamination could have moved downstream. Calculations made for resuspension of contamination in the ducting indicates that as little as  $1 \times 10^{-5}$  curies could dominate annual releases.
- KOVACH, L.: At some point, decontamination of ducts downstream of existing HEPA filter banks may be more cost effective than continued "studies". Based on currently available data, do we know what the actual release is from the stacks? Based on currently available data, do we know the DF of the existing air cleaning systems?
- **DAVIS:** The stack sampling system currently in place measures stack releases. Actual DFs are difficult to measure. However, in-place testing performed on the HEPA filters indicated that they are still performing within specifications.
- KOVACH, L.: I have some familiarity with the Hanford site, so I understand the problems. I feel that, considering the time and money being spent on evaluating things, at some point you may want to look at decontaminating the downstream side of the duct because it costs so much to maintain some of these systems. You are in a decontamination mode in most of the facilities from which these stacks are supposedly releasing activity.
- **HULL:** I cannot help but ask this question, even though it is probably not appropriate in this forum, has anybody made a cost-benefit analysis of dollars spent vs. lives saved? How much is the public paying for being protected by all this, versus the dose that they are not receiving?
- **DAVIS:** That's a very good point, because the dose from Hanford site is something like  $6x10^{-3}$  mrem/yr which is orders of magnitude below 10 mrem/yr. The regulation, 40CFR61 subpart H, requires us to evaluate it without any controls in place. This makes it necessary to upgrade a system, which is now orders of magnitude below the 10 mrem/yr limit, at a cost of about \$500,000/stack.

#### THE LINK BETWEEN

# OFF-SITE-EMERGENCY PLANNING

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#### Abstract

A variety of accident management measures has been developed and implemented in the German nuclear power plants. They constitute a fourth level of safety in the defence-in-depth concept. The containment venting system is an important example. A functioning link with well defined lines of communication between plant-internal accident management and off-site disaster emergency planning has been established.

#### 1. Introduction

Plant-internal accident management (AM) has been developed after the Chernobyl accident during the last years as a fourth additional level in the defence-in-depth concept. Its objective is to stabilize the plant in a long term safe state in cases of severe accidents which are beyond the design base incidents. A variety of AM measures both of the preventive and of the mitigative type have been developed and partially implemented in nuclear power plants (NPPs) in Germany. These measures are taken into account in probabilistic safety assessments (PSA) which represent a powerful investigative tool providing information on accident event sequence analyses and their probability of occurrence. The AM measures and their state of implementation is described in the following emphasizing the equipment for filtering and air cleaning.

In the case of an actual hazardous state developing in the plant, in parallel to the efforts to prevent core damage or to mitigate its consequences, precautionary off-site disaster control measures could be initiated. These measures and the criteria for initiation are discussed in the following.

At the end of this paper, a closer look at the link between plant-internal AM and off-site emergency planing is taken. Potential improvements in linking these two areas of high safety relevance in cases of severe accidents can be expected with respect to the source term, appropriate choice of emergency training scenarios and criteria for initiation of off-site measures.

#### 2. Accident Management Measures

# 2.1 The Defence-in-depth Concept in Nuclear Safety

Protection against possible dangers is based on two major concepts. The first consists in preventing damage by taking appropriate precautionary measures, the second tries to limit the extent of possible damages. The greater the extent of possible damages becomes, the more important become precautionary measures which, by eliminating endangering mechanisms already in the forefield, provide for a greater distance to the manifestation of the risk. In protecting essential functions, the principle of multiple defence lines in the sense of a graduated prevention has been proven to be most effective.

This led to the development of an extensive safety concept based on the principle of graduated prevention. From the beginning, one strove to develop safety measures far ahead in the preventive area wherever this was possible and reasonable from a technical standpoint. The danger potential from radioactive materials is contained within several consecutive physical barriers. These themselves are protected by a graduated system of precautionary safety measures, the defence-in-depth concept (see Figure 1).

#### - Level 1

At the first level, high quality standards are applied to all plant components with the goal of preventing failures from occurring at all. Components and systems are designed with high safety margins and subjected to extreme quality assurance measures not only during manufacture but also during the operation of the plant.

#### - Level 2

At the second level, disturbances are intercepted before they can develop into incidents. Here, the limitation system and the reactor protection system which monitors all essential measurement values of the plant are of central importance. Whenever specific limits are exceeded protective actions such as power reduction or shutdown of the reactor are initiated. The set points are specified such that overstressing of the plant components can be excluded from consideration.

#### - Level 3

Third in line is the equipment of the safety system (that means engineered safety features) which protects the radioactivity barriers in case of damage to important systems. Here the design goal is that - in case of an incident - at least two radioactivity barriers will remain intact, thus preventing a dangerous release of radioactivity to the environment. Spatial separation and special structural measures protect the safety system from damage due to events such as fire or flooding. The engineered safeguard are designed to ensure that all design basis accidents are controlled and coped with. The analytic proof of safety required for the design basis accidents will always use assumptions for insecure data or physical models which will never overestimate, rather underestimate, the effectiveness of the safety system.

#### 2.2 Accident Management Measures

The defence-in-depth concept as described in 2.1 has proven successful in establishing a high standard of safety in the nuclear field. On the other hand, experience has shown that the levels of defence not necessarily prevent accidents.

Even the most reliable safety system cannot guarantee that a system failure or a combination of failures occurs which is not covered by the design. It is for instance hypothetically possible that during a major incident the safety equipment itself fails. The analyses of beyond-design accidents, in the course of which a failure of the safety equipment is assumed, show that in most cases several hours remain before serious damage occurs to the reactor core. This leaves time for the prevention of damage by using remaining safety systems as well as operational or even external systems.

Since protective measures must be taken well in advance of failure development, it is important to apply these level-4 protective measures as early as possible within the plant and not to delay their initiation until effects of the accident on the environment become imminent.

Figure 1 shows the introduction of a fourth safety level into the defence-in-depth concept.

#### 2.3 Overview over Accident Management (AM) Measures

Accident management measures can be grouped into preventive and mitigating measures. The first group of measures is aiming at the prevention of damage to the core. The second group tries to limit the release of radioactive materials as far as possible and to prevent catastrophic effects even in the case of a severe core damage.

In Table 1, an overview is provided on accident management measures that are implemented already in German NPP or which are foreseen conceptually. Two practical examples shall be presented as applied in NPPs in Germany:

#### Coolant Injection to the Reactor Pressure Vessel (RPV) of a Boiling Water Reactor (BWR)

In the case of BWR, credit can be taken from high flexibility in the use of various injection systems for core cooling, e. g. in case of a station blackout, fire fighting systems, drinking water supply and a mobile pump can feed into the reactor pressure vessel. Emergency operating procedures ensure an activation in a relatively short time to prevent a degradation of the reactor core.

# - Containment Integrity

The containment vessel is of particular in that it presents the last physical barrier in the case of beyonddesign accidents. Therefore, accident management measures are considered for its protection almost independently from the probability of occurrence of corresponding damaging events. The measures concerned belong to the group of mitigation measures.

# Tab. 1: Overview over Accident Management Measures

# Overview over Accident Management Measures, partially implemented in German PWRs

- Emergency Manual
- Primary and Secondary Bleed and Feed
- Filtered Containment Venting
- Filtration of Air Supplied to Control Room
- System for Measuring Activity Concentration in Containment Atmosphere and Sump
- Improvement of Containment Function
- Different Measures to Improve Power Availability (Connection to Neighbouring Units, Battery Capacity, Additional Connection to Grid, Recovery of Grid Connection)

# 2.4 Air Cleaning AM-devices

Of direct relevance to nuclear air cleaning are the AM-measures filtered containment venting (FCV) and filtered air supply to the control room. Therefore, these measures shall be discussed here in more detail. The discussion is restricted to the pressurized water reactor (PWR). The objective of FCV is to prevent late overpressurization of the containment and to limit and control off-site releases. The design requirements include for the case without water injection to the containment pressure limitation at test pressure. (With water injection : Depressurization to half of the test pressure within two days). There are two different technical concepts that are applied:

- Combination of stainless-steel fibre filters for aerosol retention and venture scrubber for iodine- and additional aerosol retention
- Combination of stainless-steel fibre filters for aerosol retention with molecular sieves for iodine-retention

Existing penetrations through the containment shell are used.

The iodine filtration unit is located in the auxiliary building whereas the aerosol filter can be located inside the containment or in the auxiliary building as well.

These options are displayed in Figure 2. The possibility to arrange the aerosol filter inside the containment was achieved by progress in reducing the size of the unit. This option offers two advantages. Due to the higher pressure, the gas volume that has to be filtered is smaller. Furthermore, the radioactive aerosols filtered remain inside the containment. On the other hand, the equipment is exposed potentially to external forces caused by the accident and the filter capacity decreases with the size.

Under accident conditions, the filters of the containment venting system are exposed to extreme loads. For the case of a PWR, some typical key design data for the filter system are:

- 40 kg of aerosols to be filtered
- 2 kW decay heat of the aerosols

- decontamination factor > 1000
- vent gas temperatures in the range of 100° C to 200° C
- aerosol concentration  $\leq 5 \text{ g/m}^3$
- mass flow 3,5 kg/s

Stainless steel fibre filters have been developed in Germany to meet the requirements derived from accident analyses. These modular constructed filters consist of a first layer of metal-cloth material followed by a series of layers of stainless steel fibre filters with decreasing cross section of the fibres. Then, a moisture separator is used to reduce the steam contents that affects the final stainless steel fibre filter with 2  $\mu$ m thickness of the fibres. With the last stage decontamination factors of 10 000 for aerosols in the range of 0,1 to 0,2  $\mu$ m are achieved.

In Germany it has been decided that the control room should be the central place for planning and performing measures of accident management. In order to ensure long term habitability of the control room in case of an emergency, appropriate filter systems will be installed to limit radiation exposure from inlet air into the control room. A slight overpressure is applied to avoid inward leakage. The filter system for this purpose is usually held in reserve and installed when required.

#### 3. The Evaluation of AM Measures in Probabilistic Safety Assessment (PSA)

One of the objectives of the German Risk Studies / 3, 4 / has been the derivation and evaluation of accident management (AM) measures. AM means to make use of existing safety margins, human intervention and additional equipment in a plant for the early detection of plant conditions or sequences of events not explicitly foreseen in the design and to control these and to limit the consequences inside and outside the plant. According to the core-damage-state AM can be divided into preventive and mitigative measures / 5 /.

Examples for preventive AM measures derived from PSA-results comprise secondary and primary bleed and feed, which prevent core damage and stabilize the plant on the long term.

The AM measures are taken into account in PSAs on the basis of assumptions of the engineering judgement type concerning the corresponding success probability. It is estimated in / 3 / that AM measures reduce the frequency of uncontrollable events from  $3 \cdot 10^{-5}$  /a to  $4 \cdot 10^{-6}$  /a. The group of initiators which is mostly affected by AM measures are the transients. An impressive example for an effective AM is primary bleed and feed. The single contributions to the frequency of core damage induced by small leaks in the primary circuit or steam generator-tube rupture were reduced by two orders of magnitude, from  $10^{-6}$  /a to  $10^{-8}$  /a.

In an ongoing investigation project an evaluation concept for active and passive safety functions of the containment is under development (Containment Event Tree, CET). Essential steps during development of CET are the temporal arrangements of selected practicable AM measures in the event sequence and the limitation of the time interval of assessment from the point shortly before RPV is damaged to the loss of containment - integrity. All relevant physical phenomena during ex-vessel phase are taken into account. It is expected that effectivity of AM measures can be enhanced by the results of this investigation.

#### 4. Off-Site Disaster Control

After being informed by the plant operator of a nuclear accident, the head of the disaster control authority or his representative will decide which level of alarm (see chapter 5) shall be given. In accordance with the alarm level and the radiological condition, one or more of the following measures will be taken:

- general alert in accordance with a prescribed plan,
- identification of the endangered region,

- deployment of radiological measurement troups,
- warnings and information to the public,
- traffic restrictions,
- sheltering,
- administration of stable iodine,
- evacuation,
- alarming the drinking water supply stations,
- decontamination,
- medical care and supply.

The measures in this list that are essential to the protection of the public are the recommendations to stay sheltered, to distribute iodine tablets and to evacuate. Guidelines with respect to dose values for these recommendations are presented in the following table 2:

Reference Levels of Dose in mSv						
Countermeasure	Whole Body (external exposure and inhalation)	Thyroid (inhalation)	Lung or any other Dominantly Exposed Individual Organ (external exposure and inhalation)			
Sheltering	> 5 > 50	> 50 < 250	> 50 < 250			
Administration of Stable lodine		> 200 < 1 000				
Evacuation	< 100 < 500	> 300 < 1 500	> 300 < 1 500			

# Tab. 2: Emergency Reference Levels for Countermeasures

The dose values shown in this table are specified within individual bandwidths. This allows for a freedom of decision above the lower and below the upper dose value on executing the individual measure depending on the specific situation (e.g., depending on the extent of the emergency situation, daytime or night-time, weather condition).

A further measure for the protection of the population is the installation of emergency stations where affected persons can receive medical treatment.

A detailed discussion of off-site disaster control in Germany is provided in / 2 /. It covers the regulatory framework, the responsibilities of the Federal Government and of the States, surveillance and international obligations.

# 5. The Interface between Plant Internal Accident Management and Off-Site Emergency Measures

The link between plant internal accident management and the off-site emergency preparedness is an adequate flow of information from the plant to the concerned authorities in case of an emergency. In Germany

the operator of a nuclear power plant is bound by the nuclear licensing procedure to inform the disaster control authority when necessary. There are two alarm levels specified:

- <u>early-warning disaster alert</u> is given in case of an event in the nuclear facility where so far the effect on the environment is negligible with regard to the criteria for giving a disaster alert, where, however, on the basis of the plant condition it cannot be excluded that effects corresponding to the criteria for giving a disaster alert may develop.
- <u>disaster alert</u> is given if in case of an accident in a nuclear facility a dangerous release of radioactive materials to the environment was determined or is imminent.

The German Reactor Safety Commission (RSK) and the Radiological Protection Commission (SSK) are currently establishing specific alert criteria. These are intended to help the plant operator in deciding on the necessity for notifying the authorities and in deducing a recommendation to the authorities regarding early-warning or disaster alert from the individual accident condition. These criteria will clarify the indeterminate words in the above definitions and will allow the plant operator to differentiate between

- plant conditions due to accidents
- emissions
- immissions

in accordance with clear technical criteria or directly measured values.

Figure 3 shows a simplified scheme of the routes of information in the case of an incident or accident in a German NPP.

The operator of the NPP is obliged to inform the responsible state ministry, which is supported by a state institution performing measurements. Further state ministries have to be involved, for example the ministry which is responsible for public protection.

From the state level, the competent Federal ministry is informed (Ministry for the Environment, Nature Conservation and Nuclear Safety; BMU). BMU is responsible for communication on the international level. Support to the BMU is provided by the Federal Office for Radiation Protection (BfS) which evaluates and conducts measurements and characterizes the emergency situation radiologically.

This scheme reflects the federal structure of Germany. A more detailed description of responsibilities in the field of emergency preparedness is provided in / 2 /.

#### 6. Conclusions and Summary

In the case of an accident in a nuclear power plant (NPP), well functioning link between off-site-emergency planning and plant-internal accident management is of vital importance.

It has been outlined that accident management (AM) measures have been introduced as a fourth level of safety into the defence-in-depth concept. A considerable variety of AM measures both of the preventive and mitigative type have been implemented in German NPP, more are being prepared for implementation. Of direct relevance to nuclear air cleaning are the filtered containment venting and the filtration of air supplied to the control room. In probabilistic safety assessment (PSA) it is demonstrated that the probabilities of core melt and of large releases are substantially reduced by AM measures.

For the purposes of off-site disaster control, a number of measures can be taken by the competent disaster control authority to protect the public. Emergency reference levels of dose for countermeasures (sheltering, administration of stable iodine, evacuation) have been defined.

The link between plant-internal accident management and the off-site emergency preparedness is an adequate flow of information from the plant to the concerned authorities in case of an emergency. The operator of the NPP has to provide information to enable the authority to decide whether or not early-warning disaster alert or disaster alert is given. In Germany, work is in progress to develop specific alert criteria to facilitate decision making.

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# Figure 2: Two options for arranging perfilters and aerosol filters in pressurized water reactors



- LMU: State Ministries responsible for Environmental Protection and
- LFU: State Institution performing measurements
- BMU: Federal Ministry of the Environment, Nature Conservation and Nuclear Safety
- BfS: Federal Office for Radiation Protection
- Figure 3: Simplified flow scheme for information in the case of an incident or accident in a German NPP / 2 /

# DISCUSSION

- KOVACH, L.: Have you considered releasing stable iodine at the plant, in commensurate quantity with the released radioactive iodine, to observe the same coverage as the distribution of the radioactive fallout? You would not have to relocate all these people and you would not have to go through a very complicated administrative procedure. With the varying wind directions and everything else, I think it would be a much simpler way of administering and controlling iodine than trying to track down all the people. I am not a health physicist, so I am just wondering what would be the best way to cover everybody who would be exposed to radioactive iodine.
- **BRAUN:** I will discuss this with experts in the radiation protection commission, but I do not know whether it is possible from a technical standpoint. Too much iodine is needed as a practical reason and the public would not accept it.

#### NAUAHYGROS - A CODE FOR CALCULATING AEROSOL BEHAVIOR IN NUCLEAR POWER PLANT CONTAINMENTS FOLLOWING A SEVERE ACCIDENT

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#### Abstract

NAUAHYGROS is a computer code to calculate the behavior of fission product and other aerosol particles in the containment of a nuclear reactor following a severe accident. It is an extension of the German code NAUA, which has been in widespread use for many years. Early versions of NAUA treated various aerosol phenomena in dry atmospheres, including aerosol agglomeration, diffusion (plateout), and settling processes. Later versions added treatments of steam condensation on particles in saturated or supersaturated containment atmospheres. The importance of these condensation effects on aerosol removal rates was demonstrated in large scale simulated containment tests. The additional features incorporated in NAUAHYGROS include principally a treatment of steam condensation on hygroscopic aerosols, which can grow as a result of steam condensation even in superheated atmospheres, and improved modelling of steam condensation on the walls of the containment. The code has been validated against the LACE experiments.

# I. Introduction

NAUA (German acronym for <u>Nachunfalla</u>erosolverhaltens, postaccident aerosol behavior) is a computer code to calculate the behavior of fission product and other aerosol particles in the containment of a nuclear reactor following a severe accident. It was developed in Germany over a period of years during the 1970s and early 1980s<sup>(1)</sup>. Two current versions of the code, NAUA mod-4 and mod-5, are in widespread use, for example in the U.S. NRC Source Term Code Package, and at ENEL(Italy), VTT(Finland), and many other organizations.

Early versions of the code treated various aerosol phenomena in dry atmospheres, including aerosol agglomeration, diffusion (plateout), and settling processes. In later versions, from mod-3 on, the phenomena associated with steam condensation on particles in saturated or supersaturated steam-air atmospheres characteristic of post-accident reactor containments were treated. The importance of these condensation effects was clearly demonstrated in the MARVIKEN, DEMONA, and LACE experiments<sup>(2)</sup>. In addition mod-5 incorporated a Stefan flow diffusiophoresis model.

In recent years modelling of steam condensation effects on particles has been extended to include the effects οf hygroscopicity<sup>(3)</sup>. Hygroscopic particles can grow as a result of steam condensation even in superheated atmospheres. An improved treatment of steam condensation on interior heat sink surfaces (basically, walls) in the containment volume and its coupling to condensation on particles was developed by Li <sup>(4)</sup>, who also changed the code architecture. One of the principal changes is that NAUAHYGROS uses the MAIN program as a driver for subroutines that individually contain the models for the various aerosol (and to some extent, containment) physics phenomena. (This was done to a limited degree in mod-4.) This makes it relatively easy to add new models (e.g., thermophoresis, turbulence, sprays) that are not included in mod-4 or NAUAHYGROS. These phenomena have been left out either because they were judged to be unimportant in the context of reactor accident scenarios of interest, or because required information to handle them (e.g., detailed gas flow distributions) would probably not be available.

NAUA was never conceived as a fully coupled aerosol/thermohydraulic code. The necessary thermohydraulic data had to be obtained from another source (either experiment or a containment thermohydraulic code) and input into NAUA. In mod-4 the input thermohydraulic data consisted of the bulk atmosphere temperature, the steam mass injection rate, and the volumetric leak rate, all as functions of time. NAUAHYGROS, on the other hand, because of its more detailed treatment of condensation effects, also requires wall temperatures, containment atmosphere pressure, and possibly heat fluxes to the wall. In addition, the user must also now input certain data for hygroscopic aerosol species.

A major change has been made to the way that the aerosol source data are input. In previous versions of NAUA, two size distributions (modes) in each release period were specified, and the mass fraction of each specie in each mode was specified. In NAUAHYGROS, each specie can be released in any (arbitrary) release period with its own (log normal) size distribution. As in NAUA, the source size distribution is defined by an average particle radius and the logarithm of the geometric standard deviation.

NAUA and NAUAHYGROS are so-called "nodal point" codes, that is, the aerosol size distribution is represented as a set of discrete points or "bins", rather than by a continuous function or correlation. In addition, as will be discussed later, NAUAHYGROS is a multicomponent code with moving size bins. Briefly, "multicomponent" means that to a certain extent aerosol species are tracked individually. In the original NAUA code, all suspended aerosol species are homogeneously mixed at each time step into one average composition which is independent of size. (This is not true of the condensed water on the particles - its mass fraction is size dependent.) In NAUAHYGROS, an average composition is determined for all particles in a given size bin, but this composition varies from bin to bin. Thus, for example, if aerosol specie 1 is characterized by smaller particles than specie 2, the small size bins will have a larger fraction of specie 1 than the large bins, while the reverse situation will hold for specie 2.

"Moving" size bins arise from the fact that all particles in a given size bin will grow (or shrink) uniformly when steam condenses (or evaporates) on them, since they all have the same composition. The size of the bin can therefore be redefined after ndensation or evaporation without changing its bin identification number. Doing this insures, for example, that if one condenses steam on the particles and then reverses the situation by evaporating the steam, one will recover the original size. This is not the case in NAUA, where a particle after condensation is "split" between the bins on either side of its new size. (The splitting procedure, which was originally developed to handle particle agglomeration, is still used for that purpose in JAHYGROS.) The use of moving size bins was first described by bard<sup>(5)</sup> for the MAEROS code; it substantially reduces "numerical ffusion" effects.

#### II. Basic Theory and Modelling

#### Rate Equations

The basic aerosol equations used in NAUA (and NAUAHYGROS) will be briefly reviewed. These describe the rates at which suspended aerosol particles are added to or lost from a given size bin because of the processes of particle agglomeration (coagulation), diffusion, and gravitational settling. The particle size distribution function, which is in fact continuous, is approximated by a set of N quantities, representing the number of particles at discrete particle radius values (bins), thus forming a size histogram. The first bin corresponds to a chosen minimum radius, while the Nth bin corresponds to a chosen maximum radius. The bins are then spaced logarithmically equidistant, i.e. the ratio  $r_i / r_{i-1}$  is a constant. Clearly the larger N is, the more accurate this representation will be; on the other hand, computing time increases faster than N, approximately as  $N^2$ , (because agglomeration processes usually involve two sizes). In NAUA (and NAUAHYGROS), a value of N up to 110 can be chosen. Usually N = 30 gives sufficient accuracy with manageable computing times, although for "dry" calculations (no steam condensation on particles) larger values of N can be chosen.

It should be noted that throughout the following discussion it is assumed that the particles are spherical. Most of the applications of NAUAHYGROS are expected to involve aerosol behavior in steam/air atmospheres, in which condensation of steam on the particles will be expected to occur. It has been demonstrated<sup>(6)</sup> that under such conditions the particles will be spherical because of the surface tension of the condensed water.

User-specified shape factors to account for nonsphericity are available in NAUA and have been kept as an input option in NAUAHYGROS.

NAUA and NAUAHYGROS assume that the containment can be represented by a single volume in which the atmosphere and aerosols are uniformly distributed (well-mixed model). Thus there are no dependencies on position in the equations that follow.

In general the rate equations are of the form

$$dn_i/dt = K_i n_i \tag{1}$$

for the processes of diffusion and sedimentation, and are of the form

$$dn_i/dt = K_{ij}n_{inj}$$
 (2)

for agglomeration processes.

In equations (1) and (2)  $n_i$  is the number of particles (or the number per unit volume) in size bin i,  $K_i(r_i)$  is a rate constant characterizing the 1<sup>th</sup> removal process, and  $K_{ij}$  is an agglomeration kernel characterizing the rate of agglomeration between particles in bins i and j.

# Deposition Velocities

For the phenomena described by equation (1), it is convenient to replace the rate constants  $K_i$  by their corresponding deposition velocities  $v_i$ , where  $v_i = K_i V/A$ . A is the surface area on which the deposition is taking place (e.g., upward-facing horizontal surfaces for sedimentation), and V is the containment volume.

The  $v_i$ 's are given by the following well-known expressions:

Sedimentation:

$$v_s = 2 \rho_p g r^2 C(r) / 9\mu$$
(3)

Brownian Diffusion:

$$v_d = D(r) / \delta \tag{4}$$

Stefan Flow Diffusion (Diffusiophoresis):

$$v_{sf} = \frac{x_s \sqrt{M_s}}{(x_s \sqrt{M_s} + x_a \sqrt{M_a})} \frac{W}{\rho_s}$$
(5)

In equations (3-5)  $\rho_{\rm p}$  is the particle material density,  $\rho_{\rm S}$  is the steam density in the atmosphere, g is gravitational acceleration,  $\mu$  is the viscosity of the atmosphere, D(r) = kTC(r)/6 $\pi\mu$ r is the binary diffusion constant of the atmosphere,  $\delta$ is a diffusion boundary layer thickness, C(r) is the Cunningham slip correction factor,  $x_{\rm S}(x_{\rm a})$  is the steam (air) mole fraction in the atmosphere,  $M_{\rm S}(M_{\rm a})$  is the molecular weight of steam (air), and W is the steam condensation rate per unit area on the wall surface.

The Cunningham factor is given by

$$C(r) = 1 + 1.246 \text{ Kn} + 0.42 \text{ Kn} \exp(-0.87/\text{Kn})$$
 (6)

where Kn is the Knudsen number  $\lambda/r$ ,  $\lambda$  is the molecular mean free path in the atmosphere.

In NAUA and NAUAHYGROS, the diffusion boundary layer thickness  $\delta$  is user-specified; a value of 0.01 cm is usually recommended. The choice of  $\delta$  is not critical, since particle removal (plateout) due to Brownian diffusion is usually quite small compared with the sedimentation or diffusiophoretic deposition rates. It should be noted that the diffusiophoretic velocity is independent of particle size.

#### Agglomeration Kernels

As a result of agglomeration between particles, particles are both removed from and added to size bins. If two particles of radii  $r_1$  and  $r_2$  coagulate, the radius of the particle thus formed is  $r_3 = (r_1^3 + r_2^3)^{1/3}$ . If  $K_{12}n_{1n_2}$  is the agglomeration rate between particles of radii  $r_1$  and  $r_2$ ,

> dn1/dt = - K12n1n2 dn2/dt = - K12n1n2 dn3/dt = + K12n1n2

Note that there is a net loss of one particle per agglomeration; the total mass, however, is conserved.

Generalizing the above equations one obtains

$$dn_{i} / dt = - \sum_{j=1}^{N} K_{ij} n_{i} n_{j} + 1/2 \sum_{(j+k=i)} K_{jk} n_{j} n_{k}$$
(7)

where j+k=i means that the summation is taken over those bins for which  $r_j^3 + r_k^3 = r^3$ , where  $r^3$  includes all radii which fall between bins (i-1) and (i+1); the reason for this will become apparent later. The first term in equation (7) represents the loss out of size bin i due to coagulation with all other bins

(including coagulations between particles both of which are in bin i); the second term represents the gain in bin i due to agglomerations between smaller particles.

There are three mechanisms that lead to particle agglomeration, namely, Brownian diffusion, turbulent diffusion, and gravitational "sweepout". The first two of these result in particle-particle collisions due to the random Brownian motion or random turbulence-induced motions of the particles. Gravitational coagulation results from the fact that larger or heavier particles fall with higher (Stokes) velocities than smaller or lighter ones. The faster particles can thus overtake and collide with the slower ones in the course of their fall.

In NAUA and NAUAHYGROS only the Brownian and gravitational agglomeration processes are modeled. Turbulent agglomeration is not treated since the turbulent flow in a containment atmosphere is difficult to evaluate. The effects of electric and radiation fields are also not considered.

The agglomeration kernels are given by the following expressions:

<u>Brownian:</u>

$$K_{ij}^{B} = (4kT/6\mu) (r_{i}+r_{j}) [C(r_{i})/r_{i}+C(r_{j})/r_{j}] \qquad i \neq j$$
(8)

$$K_{ii}^{B} = (4kT/3\mu)C(r_{i}) \qquad i=j \qquad (8a)$$

These expressions are valid for particles that are large compared with the gas mean free path  $(r > 10^{-5} \text{ cm})$ .

<u>Gravitational (ri < rj):</u>

$$K_{ij}^{G} = \frac{1}{2} \frac{(r_{i})^{2}}{(r_{i} + r_{j})^{2}} (r_{i} + r_{j}) [v_{s}(r_{j}) - v_{s}(r_{i})]$$
(9)

where the term  $1/2 (r_i/(r_i+r_j))^2$  is the collection efficiency. Using equation (3), one obtains

$$\kappa_{ij}^{G} = g\rho_{p}r_{i}^{2}/9\mu \bullet [C(r_{j})r_{j}^{2} - C(r_{i})r_{i}^{2}]$$
(10)

Derivations of equation (9) are given in refs. (7) and (8). The collection efficiencies differ by a factor of three; in NAUAHYGROS the Pruppacher-Klett<sup>(8)</sup> form is used.

# Size Splitting

It was noted above that the coagulation of two particles of radii  $r_i$  and  $r_j$  will result in a particle whose radius  $r_{ij}$  is equal to  $(r_i^3 + r_j^3)^{1/3}$ . Since NAUA uses a discrete mesh of size bins, the radius  $r_{ij}$  will in general not coincide with one of the bins, but will fall between two of them. NAUA "splits" the coagulated particle.

If a coagulated particle has a radius r that falls between bins m and n, where n = m+1, the number of particles in bin m is increased by  $(r_n - r_n)/(r_n - r_m)$ , and the number in bin n is increased by  $(r - r_m)/(r_n - r_m)$ . The mass in bin m is increased by  $\frac{4\pi}{3}\rho_{\rm p}r^3(r_n-r)/(r_n-r_m)$ , while the mass in bin n is increased by  $\frac{4\pi}{3}\rho_{\rm p}r^3(r-r_m)/(r_n-r_m)$ . This procedure correctly conserves the total number of particles (one) and the total mass  $4\pi\rho_{\rm p}r^3/3$ , but does not, for example, yield a correct result for the settling rate of the coagulated particle, which is proportional to  $\rho_{\rm p}r^2$ . This is a necessary compromise in view of the finite number of discrete size bins. If the coagulated particle has a radius larger than the largest size bin, it is totally assigned to the largest bin.

The size-splitting procedure is also used in NAUA for particles whose size changes as a result of condensation or evaporation of steam. As mentioned previously, in NAUAHYGROS the moving boundary scheme makes splitting unnecessary for condensational size changes.

#### The Complete Model Equation

Combining all the processes discussed above, the following integro-differential equation results for a continuous size distribution:

$$\frac{dn(r,t)}{dt} = S(r,t) - L(r,t) - (v_s(r) + v_d(r) + v_{sf}) \frac{A}{V} n(r,t) 
+ \int_0^{\frac{r}{\sqrt{2}}} K(R',r') n(R',t) n(r',t) \frac{r^2}{(R')^2} dr' 
- n(r,t) \int_0^{\infty} K(r,r_1') n(r',t) dr'$$
(11)

where  $R' = (r^3 - r'^3)^{1/3}$ , S(r, t) is the aerosol source term, and L(r, t) (=  $\alpha_L n(r, t)$ ) represents the loss by leakage ( $\alpha_L$  is the fractional volumetric leak rate at time t).

Equation (11) must be solved numerically – for this reason the size distribution function n(r,t) is approximated by a set of discrete quantities  $n_i(r_i, t)$ , and equation (11) is replaced by:

$$\frac{dn(r_k,t)}{dt} = S(r_k,t) - (v_d(r_k) + v_s(r_k) + v_{sf} + \alpha_L(r_k))n(r_k,t)\frac{A}{V} - \sum_{i=1}^N (1 - \frac{1}{2}\delta_{ik})K(r_i,r_k)n(r_i,t)n(r_k,t) + \frac{1}{2}\sum_{i=1}^N \sum_{j=1}^N K(r_i,r_j)\beta_{ij}^k n(r_i,t)n(r_k,t)$$
(12)

where  $\delta_{ik}$  is the Kronecker  $\delta$ ;  $\delta_{ik}$ =1 if i=k,  $\delta_{ik}$ =0 otherwise.

 $\beta_{ij}^{k}$  is determined by the size-splitting formulae previously discussed if coagulation between particles in bins i and j results in a particle in bin k; otherwise it equals zero.

In NAUA there are additional terms in equations (11) and (12) that represent gains and losses due to condensational growth. These terms no longer appear in NAUAHYGROS because of the moving bins approach - it should be kept in mind that condensation does not result in a change in the number of particles in a given size bin, only a change in the radius of the particles. Nevertheless, condensational growth must still be calculated in order to determine the number and mass size distributions in terms of the new radii.

#### Condensation on Particles

The model used in mod-4 and mod-5 for condensational growth on particles is the Mason equation<sup>(9)</sup>. It can be written in the following form<sup>(8)</sup>:

$$r\frac{dr}{dt} = \frac{S - \exp(\frac{2\sigma M_w}{\rho_w RTr})}{\frac{L\rho_w}{kT} \left(\frac{LM_w}{RT} - 1\right) + \frac{\rho_w RT}{M_w D(p_{sat}(T))}}$$
(13)

where	S	= bulk steam saturation ratio
	$\sigma$	= surface tension of water on the particle or droplet
	$ ho_{\mathtt{W}}$	= density of water on the droplet
	L	= latent heat of water on the droplet
	k	= heat conductivity of water vapor
	$M_W$	= molecular weight of water (18.016 g/mol)
	D	= binary diffusion coefficient of the steam/air
		atmosphere
	Psat	= steam saturation pressure
	R	= gas constant (0.8314E8 erg/gm-deg.)
	т	= absolute temperature

From equation (13) it is apparent that for a given S greater than one (supersaturation) there is a critical radius  $r_C=2\sigma M_W/\rho_W RTln(S)$  below which condensation cannot take place. For S = 1, the critical radius is infinite, so condensation cannot take place on any size particle, while for S < 1, dr/dt is negative, so only evaporation can occur. Equation (13) is derived on the basis of various assumptions, including that rdr/dt is small compared with  $kT/\rho_W L$ . In the derivation it is also assumed that the temperature difference between the droplet and the atmosphere is small and that the steam partial pressure is small compared with the total pressure.

#### Hygroscopic Particles

For hygroscopic particles equation (13) must be modified. The exponential term in equation (13) represents the lowering of the saturation pressure at the surface of the droplet due to the curvature of the droplet, the so-called Kelvin effect. If the particle is hygroscopic or soluble, there is an additional decrease in the saturation pressure due to the solute effect. This

leads to an additional term in the numerator of equation (13):

$$r\frac{dr}{dt} = \frac{S - A \exp\left(\frac{2\sigma M_w}{\rho_w RTr}\right)}{\frac{L\rho_w}{kT}\left(\frac{LM_w}{RT} - 1\right) + \frac{\rho_w RT}{M_w Dp_{sat}(T)}}$$
(14)

where A is the water activity of the droplet aqueous solution. For dilute solutions with many hygroscopic species, the water activity can be determined by the following equation <sup>(3)</sup>:

$$A = \sum_{i} \frac{1}{1 + Q_{i} M_{w} m_{i}}$$
(15)

where  $Q_i$  is the van't Hoff factor and  $m_i$  is the molality of each hygroscopic material dissolved in the droplet.

In the analysi's of the growth of hygroscopic particles it is important to know the chemical composition of these particles. because the hygroscopic growth of particles depends strongly on the amount and properties of water soluble matter in the particle. Most inorganic salts, e.g. CsI, when exposed to increasing humidity, will dissolve at a certain relative humidity (deliquescent point) which depends on the properties of the electrolyte and the system temperature. At the deliquescent point particles which contain these compounds will form saturated droplets and undergo at the same time an abrupt increase in size. As the humidity increases further, the droplets will grow and become more dilute. When the humidity decreases, the droplets become smaller and the particles recrystallize at a relative humidity (RH) considerably lower than their deliquescent point. In the case where particles also contain insoluble compounds, these two humidities are near each other. On the other hand, a few electrolytes, such as CsOH and NaOH, will form an aqueous solution and start to absorb water at a very low RH (< 10 %). Particles consisting of these substances grow smoothly with increasing RH.

Equation (14) has a number of interesting features. First, there is still a critical radius, but all particles larger than this critical radius will have an equilibrium radius when RH is less than one. This equilibrium radius is reached very rapidly when RH is less than about 0.99. In NAUAHYGROS immediate equilibrium is assumed when RH is less than 0.99. Also the rate at which the particles grow is not very critical, because the final size at given RH is determined by the equilibrium radius and not by the rate steam is condensing on the particles. In contrast where only nonhygroscopic particles are present there is no equilibrium radius, and it is the condensation rate which determines the particle size.

Equation (15) is valid only for dilute solutions (i.e. high RH) and it will give conservative (too large) values at low RH. However, this is not important in reactor accident applications, because at low RH even hygroscopic particles do not grow much.

The parameters that are required to evaluate the water activity include the van't Hoff factor and the mole fraction of each hygroscopic material in the particle (equation (15)). In addition the minimum relative humidity at which deliquescence occurs must be specified. The van't Hoff factor and minimum relative humidity are user-input. Mole fractions are updated by the code at each time step. The water activity is also a function of temperature and concentration. For a given initial dry mass of hygroscopic material, the concentration will change as water condenses on the particle, thus A is continuously changing (except at the equilibrium size or concentration).

#### Wall Condensation Models

As shown above, condensation on particles is driven by the relative humidity (saturation ratio) in the containment atmosphere. To calculate the relative humidity and update it at each time step, NAUAHYGROS tracks the mass balance of both water and steam, and combines this information with the current atmosphere temperature and pressure. A correct evaluation of the relative humidity can only be done if condensation of steam on the containment wall is taken into account. In addition the Stefan flow diffusiophoretic effect on particle deposition depends directly on the wall condensation rate (equation (5)).

Calculation of condensation on a wall is an old problem in heat transfer and fluid mechanics, and a number of theoretical models and engineering correlations have been developed to handle it. In mod-4 there is no wall condensation model. In mod-5 a simple approach is taken: the wall condensation rate is set equal to the steam injection rate, and the saturation ratio then varies only because of condensation on the particles and changes in atmosphere temperature. In NAUAHYGROS, as has been mentioned, the wall condensation and particle condensation rates are calculated independently, as functions of the saturation ratio or other thermohydraulic quantities at the end of the preceding time step, then the saturation ratio is recalculated (updated) for use in the next time step, using mass balance to determine the new steam density in the atmosphere.

Four models are available in NAUAHYGROS for the wall condensation calculation. The user selects one by the choice of an input parameter MODEL (= 1-4).

# Combined Nusselt-Heat Transfer/Mass Transfer Analogy

MODEL = 1 selects a combination of the classical Nusselt equation<sup>(10)</sup> on the liquid (condensate) side and the well-known equation that results from the heat transfer/mass transfer analogy<sup>(11)</sup> on the vapor side. The Nusselt equation is:

$$W = \frac{4}{3} \left\{ \frac{k(T_{gas} - T_f)}{L} \right\}^{\frac{3}{4}} \bullet \left\{ \frac{\rho_w(\rho_w - \rho_s)}{4\mu_w H} \right\}^{\frac{1}{4}}$$
(16)

The heat transfer/mass transfer analogy equation is:

$$W = \frac{DM_w p}{RT_f \delta_v p_{an}} (p_{ai} - p_{a\infty})$$
<sup>(17)</sup>

In equations (16) and (17), in addition to previously defined symbols,

$^{T}f$	=	condensate film surface temperature
$\mu_W$	=	viscosity of liquid water
$ ho_{{ m s}}$	=	density of steam
Н	=	height of the wall
р	=	total pressure in the containment atmosphere
р <sub>аі</sub>	=	air partial pressure at the condensate surface
Pam	=	logarithmic mean of the air partial pressure through the boundary layer at the condensate surface
Pa∞	=	air partial pressure in the bulk atmosphere
$\delta_{ m V}$	=	mass transfer boundary layer thickness

 $\delta_{\rm V}$  is calculated according to the convective regime (forced or natural) as a function of the Grashof and Schmidt numbers, which are internally calculated.

The program solves equations (16) and (17) iteratively to find a value of the condensate film surface temperature  $T_{\rm f}$  that makes the two condensation rates equal.

This method is used in a number of reactor and severe accident thermohydraulic codes such as MAAP, MELCOR, and CONTAIN. It is therefore the recommended choice when the input thermohydraulic data to NAUAHYGROS are obtained from calculations using these or similar codes.

#### Combined Nusselt - Kinetic Theory Model

MODEL = 2 selects the Nusselt - Kinetic Theory model<sup>(12)</sup>. This model has been shown<sup>(4)</sup> to give the best overall agreement with aerosol and condensation data from the LACE experiments<sup>(13)</sup>. The model combines the Nusselt equation (16) with the following equation derived from kinetic theory considerations<sup>(4,8,14)</sup>

$$W = \frac{\alpha_c}{(1 - 0.5\alpha_c)} \left(\frac{M_w}{2\pi RT_w}\right)^{\frac{1}{2}} p_{sal}(T_w) \left[S - \frac{p_{sal}(T_f)}{p_{sal}(T_w)}\right]$$
(18)

where  $S = p_S/p_{sat}(T_{\infty})$ ,  $p_S$  is the steam partial pressure at the condensate surface (assumed equal to the bulk steam partial pressure),  $p_{sat}(T_f)$  is the steam saturation pressure at the surface temperature of the condensate film, and  $p_{sat}(T_{\infty})$  is the bulk steam saturation pressure.

As in model 1, the film surface temperature that gives equal condensation rates from equations (16) and (18) is found by iteration. In these models the vapor side condensation rate is driven by the relative humidity S. (Equation (17) can also be cast into a form similar to equation (18) if the steam partial pressure is small compared with the total pressure.)

In equation (18) the constant  $\alpha_{\rm C}$  is the so-called condensation coefficient. Its value is somewhat uncertain. It has been measured for both "renewing" and "non-renewing" surfaces by a number of investigators <sup>(8)</sup>. For renewing surfaces

the experimental values tend to be close to one; for non-renewing surfaces they are much smaller. A thin condensate film represents a n-renewing situation (there is no fresh supply of water from deep in the condensate film), thus a value of 0.036 has been chosen for NAUAHYGROS as a reasonable average of the experimental values. However, condensation rates calculated in NAUAHYGROS are quite insensitive to the value of  $\alpha_{\rm C}$ , since the relative humidity "adjusts" to compensate for the difference in  $\alpha_{\rm C}^{(4)}$ .

One of the basic assumptions underlying equation (18) is that the speam partial pressure is constant right up to the condensate surface, i.e., S in equation (18) refers to the bulk steam partial pressure and there are no boundary layer effects. This might be expected to be the case in the presence of turbulence, and is probably the reason that this model gives the best agreement with experimental data, as mentioned earlier. It is recommended that

this model be used when experimental thermohydraulic data are supplied.

#### <u>Uchida - Tagami Correlation</u>

MODEL = 3 employs the Uchida - Tagami correlation<sup>(15,16)</sup>. The Uchida and Tagami correlations are quite similar, and the Tagami form is used in NAUAHYGROS. The correlation gives the heat transfer coefficient to the wall, h, in terms of the steam-to-air mass ratio,  $f_V$ . Multiplication by the temperature difference  $(T_{\infty}-T_W)$  and division by the latent heat of condensation yields the mass condensation rate. The correlation is:

$$h = 11.4 + 284(\frac{f_v}{1 - f_v}) \tag{19}$$

The units are SI; NAUAHYGROS internally makes the conversion to cgs units.

This choice is recommended when the source of the input thermohydraulic data is a code that uses it.

#### Direct Use of Input Heat Flux Data

MODEL = 4 uses input heat fluxes to the wall to directly infer the wall condensation rate (both models 3 and 4 assume that sensible heat transfer to the wall is negligible compared with condensation heat transfer).

This model is recommended when heat transfer rates are available, either from experiment or a code calculation. The input heat fluxes must be in cgs units. If condensation rates are available from an external source, they can be used by converting them to heat fluxes, which can then be input. The choice of MODEL = 4 will then recover the condensation rates directly.

#### Stefan Flow Diffusiophoresis

Equation (5) gives the diffusiophoretic deposition velocity in terms of the wall condensation rate. NAUAHYGROS calculates the condensation rate W and the steam density and mole fraction at each time step, using the chosen wall condensation model to obtain W.

#### III., Validation of NAUAHYGROS

The principal validation of NAUAHYGROS has been against the LACE experiments LA-2, LA-4, and LA- $6^{(13)}$ . These tests were performed in a large (850 m<sup>3</sup>) tank. Hygroscopic (CsOH) and non-hygroscopic (MnO) aerosols were introduced over an approximately

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one hour period at a rate that resulted in maximum aerosol concentrations of 1-10  $g/m^3$ , similar to expected accident concentrations. Steam was also injected so as to provide saturated or near-saturated conditions. (The steam and aerosol injection protocols and leakage conditions were somewhat different in the three tests). The tests were well instrumented to measure aerosol parameters such as suspended aerosol mass as a function of time, integrated settled, plated, and leaked mass, and size distribution parameters. In addition, extensive thermal hydraulic measurements were made, which provided benchmark and input data for the code calculations.

Figure 1 shows the measured and calculated suspended mass for the three tests. It should be noted that at late times, when the suspended aerosol concentrations are of the order of two orders of magnitude or more below their peak values, the calculated and measured concentrations can disagree considerably. However, the impact of this disagreement on integrated aerosol quantities is negligible.

Table 1 shows the integral data for tests LA-2 and LA-4 (integral data for LA-6 were not taken). It should be emphasized again that calculations that do not properly account for diffusiophoresis and steam condensation on the particles fail quite badly in predicting the aerosol behavior in these experiments. (Thermophoresis was not significant in these tests). On the other hand these effects might not be as important in other situations, depending on the thermal hydraulic conditions.

There is a slight inconsistency between the measured suspended mass concentrations in LA-2 shown in Figure 1 and the measured integrated leakage for this test. The leakage derived from the measured suspended mass data agrees better with the NAUAHYGROS results than the measured leakage does.

Table 1.	Integrated settled,	plated, and	leaked aerosol,
	Tests LA-2	AND LA-4	

	Settled (g)	Plated (g)	Leaked (g)
LA-2, test data	1973(±10%)	449(±20%)	1515(±15%)
LA-2, calculated	2366	334	1197
LA-4, test data	4490(±10%)	532(±20%)	108(±30%)
LA-4, calculated	4437	609	96
LA-4, calc. w/o steam	4811	10	321



LA-2 NAUAHYGROS calculation, line



5







Standard deviations on the data points range from  $\pm 5\%$  to  $\pm 60\%$ .



#### IV. Summary

NAUAHYGROS is a code that simulates aerosol behavior in a nuclear power plant containment following a severe accident. It is an extension of the much used code NAUA that includes several important improvements in both the modelling of the aerosol behavior and the code architecture. Among the principal new features of the code are the ability to treat steam condensation effects, both on the aerosol particles (including hygroscopic species) and on internal heat sink surfaces, e.g. the walls of the containment; the ability to introduce up to 50 aerosol species at arbitrary times and with arbitrary (lognormal) size distributions; and a "moving bin" feature that substantially eliminates numerical diffusion effects in the treatment of steam condensation on the particles. Most of the important aerosol removal mechanisms are modelled; those that are not, e.g. thermophoresis and deposition mechanisms due to turbulence, can be readily added as new subroutines.

The code has been validated against the LACE containment experiments, giving satifactory agreement with suspended mass concentrations, integrated settled, plated, and leaked masses, and other aerosol properties (e.g. ammd). The inclusion of steam condensation effects, particularly hygroscopicity, was essential in order to obtain this agreement.

A more complete description of the code is given in ref.17, which is also a user's manual.

#### Acknowlegements

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#### DISCUSSION

- WREN: Are the temperatures of walls and gas phase kept constant with time in the code? Does the aerosol behavior affect these temperatures with time? If so, how can these changes be modelled?
- SHER: Temperatures are not constant they are input data. NAUA (or NAUAHYGROS) does not calculate the effect of aerosols on temperatures. T/H codes, which can be the source of the temperature tables, may do so. There is no code that fully couples aerosols and T/H behavior.

# 23<sup>rd</sup> DOE/NRC NUCLEAR AIR CLEANING AND TREATMENT CONFERENCE

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# Experimental study on the particles deposition in the sampling duct

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# Abstract

A high standard of protection against the harmful effects of radioactive aerosol dissemination requires a measurement, as representative as possible, of their concentration. This measurement depends on the techniques used for aerosol sampling and transfer to the detector, as well as on the location of the latter with respect to the potential sources. The aeraulic design of the apparatus is also an important factor.

Once collected the aerosol particles often have to travel through a variably shaped duct to the measurement apparatus. This transport is responsible for losses due to the particles deposition on the walls, leading to a distortion on the concentration measurements and a change in the particle size distribution.

To estimate and minimize measurement errors it is important to determine the optimal transport conditions when designing a duct; its diameter and material, the radius of curvature of the bends and the flow conditions must be defined in particular.

This paper presents an experimental study in order to determine, for each deposition mechanism, the retained fraction, or the deposition velocity for different flow regimes.

This study has pointed out that it exists a favourable flow regime for the particle transport through the sampling ducts (2500 < Re < 5000). It has been established, for any particle diameters, equations to predict the aerosol penetration in smooth-walled cylindrical metal ducts.

# I. Introduction

A high standard of protection against the harmful effects of radioactive aerosol dissemination requires a measurement as representative as possible of their concentration. This measurement is often realised after a transport, in a variably shaped duct, of the collected particles. The transport is responsible for losses due to the particles deposition on the walls, leading to a distortion on the concentration measurement and a change in the particle size distribution.

The deposition mechanisms of the particles in a duct are more or less important according to the particle size and flow regimes. The main mechanisms considered are as follows :

- brownian diffusion
- sedimentation
  - impaction
  - turbulent diffusion
  - electrostatic precipitation

The effect of each mechanism can be quantified by the retained fraction  $F_R$ , expressed by the ratio between the concentrations measured downstream (C) and upstream (C<sub>o</sub>) from the duct, i.e.:

$$F_{R} = 1 - \frac{C}{C_{o}}$$

In laminar flow, for each case considered, an expression exists for the calculation of the retained fraction  $F_{R}$ .

In turbulent flow we use the notion of deposition velocity, K, considered normal and directed towards the wall.

$$\mathbf{K} = \frac{\mathbf{n}}{\mathbf{C}}$$

with : n : deposition rate C : average aerosol concentration

The fraction hold back is then given by the general expression :

$$F_{R} = 1 - \exp\left(-\frac{aKL}{\overline{U}D}\right)$$

with :

L : pipe length

D : pipe diameter

U: mean flow velocity

a = 4: deposition on the entire surface of the duct

a = 2: deposition on a half-surface of the duct

This paper proposed some expressions for the retained fraction  $F_R$  or the deposition velocity K for each of the mechanisms considered and for different flow conditions. These expressions are derived from an experimental study [1] conducted on a cylindrical experimental duct made up of 8 assembled metallic elements. The use of monodispersed fluorescein aerosols allows a detailed measurement of the retained fraction which is determined by washing of the downstream elements and filter and by fluorimetric measurement of the solutions obtained. Certain elements may be replaced by tubes with non-conducting walls or of different diameters. The test circuit uses, to produce monodispersed particles, a Differential Mobility Analyser (DMA), in the range 0.01 to 1  $\mu$ m and a Vibrating Orifice Generator (VOG) in the range 1  $\mu$ m to 10  $\mu$ m. The aerosol electric charge, measured by an electrometric system, is varied by ionising and neutralising systems. A bubbler gives a humidity variation of about 10 to 50 % R.H. The air is pumped through the circuit at flow rates corresponding to Reynolds numbers ranging from 0 to 10 000 for a pipe diameter of D = 2.8 cm.

# II. Deposition mechanisms

# Brownian diffusion

Fine particles, like gas molecules, are subject to Brownian movement owing to their thermal agitation, the particles can strike the wall of the duct and remain fixed.

# ► Laminar flow

The fraction retained in a duct of length L may be calculated from the equations established by Townsend, Gormley, Kennedy, Demarcus [2].

If 
$$\alpha \ge 0.01$$
;  $F_R = 1 - 0.819 \exp(-3.657 \alpha) - 0.097 \exp(-22.3 \alpha) - 0.032 \exp(-57 \alpha)$   
If  $\alpha < 0.01$ ;  $F_R = 2.56 \alpha^{2/3} - 1.2 \alpha - 0.177 \alpha^{4/3}$ .

with : 
$$\alpha = \frac{\pi \text{ Di } L}{Q}$$
 Di =  $\frac{TC}{3 \pi \mu \text{ dp}}$ 

- Di : aerosol diffusion coefficient
- Q : flow rate
- dp : particle diameter
- $\mu$ : dynamic viscosity
- C: Cunningham factor
- K: Boltzmann constant

# ► Turbulent flow

If the diffusion of fine particles is assumed to take place through the laminar sub-layer of thickness,  $\delta$ , and if the concentration gradient in this sub-layer is assumed linear, we get :

$$\mathbf{K} = \frac{\mathbf{D}\mathbf{i}}{\mathbf{\delta}}$$

with  $\delta = A \cdot \frac{v}{U_*}$ , where 5 < A < 11.5 depending the author concerned, hence

U.: friction velocity : 
$$U_* = \overline{U} \left(\frac{f}{2}\right)^{1/2}$$

f : friction factor :  $f = \frac{0.316}{4 (Re)^{0.25}}$  for smooth turbulent flow

 $\nu$ : kinematic viscosity

A single equation, relating dimensionless values, is derived from the experimental results :

$$K_{\perp} = 0.123 \text{ Sc}^{-3/4}$$

with :

Sc = 
$$\frac{v}{Di}$$
: Schmidt number  
 $\frac{K}{U_{\star}}$ : dimensionless deposition velocity

The experiments were conducted on a metallic vertical tube (D = 2.8 cm) with monodispersed aerosols (0.025  $\mu$ m < dp < 1.1  $\mu$ m) in different flow conditions (3 300 < Re 10 000).

From these experiments, the dimensionless deposition velocity is calculated and ploted versus Schmidt number in figure 1. In this figure the Wells and Chamberlain equation ( $K_{+} = 0.2 \text{ Sc}^{-2/3}$ ) is also represented.

The retained fraction  $F_R$  is determined from K and a = 4 with the general expression given in introduction.

# Deposition due to combined gravitational and centrifugal forces

In the case of an horizontal bend of curvature radius R, a single expression of K may be written, accounting for the relative accelerations of gravitational and centrifugal forces :

$$K = \frac{2}{\pi} \cdot \tau \left[ \frac{g^2 + \left(\frac{\overline{U}^2}{R}\right)^2}{g + \frac{\overline{U}^2}{R}} \right]$$

The case of the vertical bend is more difficult to estimate since the angle between the vectors of the forces varies with the position of the particle in the bend, whereas this value is always  $\pi/2$  when the bend is horizontal.

# Turbulent diffusion

The gas eddies of a turbulent flow may cause certain particles to be projected towards the wall through the boundary layer. This turbulent diffusion is independent of the orientation of the duct and can sometimes be the only phenomenon responsible for deposition in vertical tubes.

Liu et Agarwal [4] have given an expression for the dimensionless deposition velocity :

if 
$$\tau_+ < 15 \ \mathrm{K_+} = 6.10^4 \ \tau_+^2$$

if 
$$\tau_+ \ge 15 \ \mathrm{K}_+ = 0.13$$

where  $\tau_+$ : dimensionless relaxation time :  $\tau_+ = \tau \frac{U_+^2}{v}$ 

Experiments were developed in a metallic vertical tube (D = 2.8 cm) with monodispersed aerosols  $(0.6 \ \mu m < dp < 9 \ \mu m)$  in different turbulent flow conditions (3 400 < Re < 10 000). Dimensionless deposition velocity deduced from these experimental results are presented in the figure 2 and compared with the expression given by Liu and Agarwal. This results pointed out a lower limit for this expression when  $\tau_+ < 0.1$ .

# Deposition of charged particles due to the image force

We know the velocity of attraction, due to the image force, of a particle of charge p.e at a distance y from a wall likened to a plane :

$$\frac{dy}{dt} = \frac{-\text{ Di } (\text{p.e})^2}{\text{KT } 4y^2} \cdot \frac{1}{4 \pi \epsilon_o}$$

This velocity, normal to the wall, is the deposition velocity.

Tests carried out for variable flow conditions  $1\ 200 < \text{Re} < 10\ 000$  have shown that deposition is minimal at the beginning of turbulent flow, Re ~ 2 500, then varies with the thickness of the laminar sub-layer  $\delta$ .

If we consider the particles to undergo a constant attraction velocity at distance  $y = \delta$  from the wall, we get :

$$K = \frac{\text{Di. } (\text{pe})^2}{\text{T 4 } \delta^2} \cdot \frac{1}{4 \pi \epsilon_0}$$

Tests with 0.6 to 9  $\mu$ m particles diameters of variable charge and for variable Re have yielded an approximate  $\delta$  value :

$$\delta = 2.2 \frac{v}{U_*}$$

This distance is inside the laminar sub-layer, of thickness :

$$5 \frac{v}{U_*} < \delta < 11.5 \frac{v}{U_*}$$

# III. Experimental studies

# ▶ Influence of the Reynolds number

The variation of the particle deposition in any element of a complex circuit were studied for different flow conditions and particle sizes in tubes of 2.8 cm in diameter.

Figures 3, 4 and 5 plot the variations in the 4  $\mu$ m and 9  $\mu$ m particle fractions held back in the following elements :

- Straight horizontal tube : in laminar flow the measurements verify the NATANSON equation since Re = 2500; in turbulent flow condition, the experiments show that the Fuchs equation is not verified because it is necessary to take into account the effects of the turbulent diffusion.
- Straight vertical tube : the measured deposits, due entirely to turbulent diffusion, line up quite well with the Liu and Agarwal equation.
- Horizontal and vertical bends for which the radius of curvature is equal to 8 times the pipe diameter; the fraction retained is obviously greater in the horizontal bends and shows in both cases, vertical and horizontal, a minimum when the Reynolds number is around 3 000.

Figure 6 expresses the fraction retained in the whole experimental circuit which is composed of pipes of 2.8 cm in diameter with horizontal linear tube of 6 m long, vertical linear tube of 2.2 m long, two horizontal bends of 90° angle and two vertical bends of 90° angle. This figure shows that it exists for the largest particles a favourable flow conditions around Re = 3500 to reduce the particle deposition.

# ▶ Influence of the particle diameter

The curve of figure 7, corresponding to the entire complex circuit, reveals a minimum deposition around 0.25  $\mu$ m. As the particle size increases from 0.01 to 10  $\mu$ m the molecular regime gives way to the inertial regime, the particle are less and less sensitive to the Brownian motion as their weight increases, while gravitational and centrifugal forces and that of diffusion due to flow eddies become stronger.

# IV. Conclusion

Any transport gives rise to losses, so ducts should only be used when absolutely necessary. In such cases, the loss of aerosol can be reduced to a minimum.

As a general rule, the deposition in a transport duct involving elements of different shapes (straight, curved) and orientations (horizontal, vertical) is least when the flow is slightly turbulent : 2 500 < Re < 5 000.

This applies especially to large particles, and is also a factor in reducing deposition due the electric charge (image force) of aerosols. For the finest aerosol particles, it is safe to use a more turbulent flow, the deposition rate decreasing slowly as the Reynolds number rises. When the tube diameter is small (D < 20 mm) it is essential to choose a flow rate such that Re = 2 500 in order to avoid turbulent diffusion, which would then become a very highly influential factor.

It is advisible to use as large pipe diameter as possible while keeping the favourable flow conditions and reduce as much as possible the horizontal tubes and curves. The bends must have a radius of curvature greater than 8 times the duct diameter and to minimize electrical effects it is better to use metallic tubes electrically conducting and earthed, because the triboelectrical effect on nonconductive walls induces very high retention of charged particles.

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# DISCUSSION

SHER: Were these experiments done dry, no steam?

- **VENDEL**: Experiments were made in dry conditions and with variable relative humidity.
- **SHER**: What was the aerosol?
- **VENDEL**: The aerosol was sodium fluorecein.
- SHER: In the LACE experiments, they did retention measurements in long pipes, and found that the results varied considerably, from a few percent to as much as 90% retention, depending on the nature of the aerosol (composition, hygroscopicity). I wonder if you observed anything like that. These experiments were in steam conditions, i.e., high relative humidity.
- **VENDEL:** Retention does not depend strongly on the nature of the aerosol when the conditions are dry air. In the case of hygroscopic particles in wet conditions, the rate of deposition can be affected, because the particle sizes vary according to the retention time, and the rate of deposition must be evaluated during the entire time.

# CLOSING COMMENTS OF SESSION CO-CHAIRMAN BELLAMY

In this session we have learned where the NRC stands with its revised accident source terms. We have heard a little bit about the effect of EPA regulations on monitoring needs at the Westinghouse Hanford facility. We have discussed emergency planning in Germany, aerosol behavior inside containments after severe accidents, and, finally, some concerns about particle deposition inside sampling ducts. We want to thank the presenters and the commenters for their interest this morning.