Organisation de Coopération et de Développement Economiques Organisation for Economic Co-operation and Development

OLIS : 29-Mar-2000 Dist. : 30-Mar-2000

English text only

PARIS

NEA/CSNI/R(2000)9

Unclassified

NUCLEAR ENERGY AGENCY COMMITTEE ON THE SAFETY OF NUCLEAR INSTALLATIONS

INSIGHTS INTO THE CONTROL OF THE RELEASE OF IODINE, CESIUM, STRONTIUM AND OTHER FISSION PRODUCTS IN THE CONTAINMENT BY SEVERE ACCIDENT MANAGEMENT

English text only

ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

Pursuant to Article 1 of the Convention signed in Paris on 14th December 1960, and which came into force on 30th September 1961, the Organisation for Economic Co-operation and Development (OECD) shall promote policies designed:

- to achieve the highest sustainable economic growth and employment and a rising standard of living in Member countries, while maintaining financial stability, and thus to contribute to the development of the world economy;
- to contribute to sound economic expansion in Member as well as non-member countries in the process of economic development; and
- to contribute to the expansion of world trade on a multilateral, non-discriminatory basis in accordance with international obligations.

The original Member countries of the OECD are Austria, Belgium, Canada, Denmark, France, Germany, Greece, Iceland, Ireland, Italy, Luxembourg, the Netherlands, Norway, Portugal, Spain, Sweden, Switzerland, Turkey, the United Kingdom and the United States. The following countries became Members subsequently through accession at the dates indicated hereafter: Japan (28th April 1964), Finland (28th January 1969), Australia (7th June 1971), New Zealand (29th May 1973), Mexico (18th May 1994), the Czech Republic (21st December 1995), Hungary (7th May 1996), Poland (22nd November 1996) and the Republic of Korea (12th December 1996). The Commission of the European Communities takes part in the work of the OECD (Article 13 of the OECD Convention).

NUCLEAR ENERGY AGENCY

The OECD Nuclear Energy Agency (NEA) was established on 1st February 1958 under the name of the OEEC European Nuclear Energy Agency. It received its present designation on 20th April 1972, when Japan became its first non-European full Member. NEA membership today consists of 27 OECD Member countries: Australia, Austria, Belgium, Canada, Czech Republic, Denmark, Finland, France, Germany, Greece, Hungary, Iceland, Ireland, Italy, Japan, Luxembourg, Mexico, the Netherlands, Norway, Portugal, Republic of Korea, Spain, Sweden, Switzerland, Turkey, the United Kingdom and the United States. The Commission of the European Communities also takes part in the work of the Agency.

The mission of the NEA is:

- to assist its Member countries in maintaining and further developing, through international co-operation, the scientific, technological and legal bases required for a safe, environmentally friendly and economical use of nuclear energy for peaceful purposes, as well as
- to provide authoritative assessments and to forge common understandings on key issues, as input to government
 decisions on nuclear energy policy and to broader OECD policy analyses in areas such as energy and sustainable
 development.

Specific areas of competence of the NEA include safety and regulation of nuclear activities, radioactive waste management, radiological protection, nuclear science, economic and technical analyses of the nuclear fuel cycle, nuclear law and liability, and public information. The NEA Data Bank provides nuclear data and computer program services for participating countries.

In these and related tasks, the NEA works in close collaboration with the International Atomic Energy Agency in Vienna, with which it has a Co-operation Agreement, as well as with other international organisations in the nuclear field.

CSNI

The NEA Committee on the Safety of Nuclear Installations (CSNI) is an international committee made up of senior scientists and engineers, with broad responsibilities for safety technology and research programmes, and representatives from regulatory authorities. It was set up in 1973 to develop and co-ordinate the activities of the NEA concerning the technical aspects of the design, construction and operation of nuclear installations insofar as they affect the safety of such installations. The Committee's purpose is to foster international co-operation in nuclear safety amongst the OECD Member countries. CSNI's main tasks are to exchange technical information and to promote collaboration between research, development, engineering and regulation organisations; to review the state of knowledge on selected topics of nuclear safety technology and safety assessments, including operating experience; to initiate and conduct programmes to overcome discrepancies, develop improvements and reach consensus on technical issues; to promote co-ordination of work, including the establishment of joint undertakings.

PWG4

CSNI's Principal Working Group on the Confinement of Accidental Radioactive Releases (PWG4) has been given two tasks: containment protection, and fission product retention. Its role is to exchange information on national and international activities in the areas of severe accident phenomena in the containment, fission product phenomena in the primary circuit and the containment, and containment aspects of severe accident management. PWG4 discusses technical issues/reports and their implications, and the results of International Standard Problem (ISP) exercises and specialist meetings, and submits conclusions to the CSNI. It prepares Technical Opinion Papers on major issues. It reviews the main orientations, future trends, emerging issues, co-ordination and interface with other groups in the field of confinement of accidental radioactive releases, identifies necessary activities, and proposes a programme of work to the CSNI.

FPC

The Task Group on Fission Product Phenomena in the Primary Circuit and the Containment (FPC) is a specialised extension of PWG4. Its main tasks are to exchange information, discuss results and programmes, write state-of-the-art reports, organise specialist workshops, perform ISPs in the field of fission product phenomenology.

CAM

The Task Group on Containment Aspects of Severe Accident Management (CAM) is a specialised extension of PWG4. Its main tasks are to exchange information, discuss results and programmes, write state-of-the-art reports, organise specialist workshops on containment accident management and on techniques to protect the containment and their implementation.



FOREWORD

CSNI's Principal Working Group on the Confinement of Accidental Radioactive Releases (PWG4) had decided the preparation of a Technical Note on accident management aspects of the control of the release of iodine, cesium, strontium and other fission products in the containment during a severe accident. The original purpose was to prepare a report based on the following outline:

- a brief discussion of fission product sources; fission product characteristics; chemical compounds;
- transport and deposition of fission products; brief description of different deposition and agglomeration processes;
- retention of fission products; re-evaporation, resuspension, etc.;
- discussion of various possibilities to enhance the removal of fission products from the containment atmosphere;
- discussion of various possibilities to retain the fission products in the sumps, on surfaces or in filters;
- discussion of the possible measures.

The first three items had been described in a fair amount of detail in other documents. It was agreed that PWG4's Task Group on Fission Product Phenomena in the Primary Circuit and the Containment (FPC) would be responsible for preparing this part of the report. Dr. Dana Powers (Sandia National Laboratories, USA) took the lead on this work, with the assistance of FPC members. We are most grateful for the considerable time and effort he spent on this, translating complex knowledge into legible language, and producing an excellent overview of the technical bases for the management of severe reactor accident source terms.

In a second phase, members of PWG4's task Group on Containment Aspects of Severe Accident Management (CAM) "enriched" the report with severe accident management considerations. These can be found at the end of Chapters III to VIII.

The report does not aim at exhaustiveness, nor at direct applicability to severe accident management situations. Its aim is to give a picture of what is known in the area of fission product sources and how this knowledge can be put to use to mitigate them. Implementation has to be developed on a plant-by-plant basis, taking account of plant specifics. This is the task of the utilities.



PROLOGUE

This document is intended to provide a management-level overview of the technical bases for accident management activities to attenuate releases of radioactive materials in the very unlikely event of a severe nuclear power reactor accident - activities known commonly as management of severe accident source terms. Such activities are natural complements to accident management activities directed at arresting or slowing accident progression. Abbreviated, qualitative discussions are presented in the document on the more important severe nuclear reactor accidents, the nature of radioactive material releases during accidents, natural processes that act to attenuate the amount of radioactive material that can escape a power plant, and the physical and chemical principles used in engineered systems to further attenuate radioactive releases during accidents.

At the end of each section of the report, an annotated bibliography is provided. These bibliographies are intended to serve as introductions to the vast literature pertinent to all aspects of accident management including the management of radioactive source terms. The bibliographies are by no means comprehensive. Literature has been selected for citation largely because the authors found it useful to their understanding of the technical issues. Many good and even better works have not been cited simply to keep the length of the report manageable. An effort has been made to cite the proceedings of major conferences that contain many fine contributions, which provide the interested reader with a broad view of the worldwide effort in nuclear reactor safety. The authors apologize to those whose work has not been cited explicitly. It is also true that the field continues to evolve and the literature of the field continues to improve. It is simply not possible to keep the bibliographies up to date as new works continue to appear. The cited literature has been chosen to reflect that state of the field of source term accident management up to about 1997. It is hoped that the bibliographies are sufficient to provide the interested reader with information to locate the many other useful reports and papers as well as to keep abreast of the continuing developments in the field of source term accident management.

Finally, it must be noted that much of the presentation has been made from the perspective of conventional pressurized water reactors and boiling water reactors. Many important details will be different for other types of reactors or for reactors with special features. Readers are asked to do the mental manipulations necessary to apply the ideas discussed here to the particular circumstances and features of their own reactors.

TABLE OF CONTENTS

I.	INTI	RODUCTION	11		
	I.1	Accident management	11		
	I.2	Annotated bibliography on accident management	13		
II.	THE	THE SOURCES			
	II.1	Introduction	15		
	II.2	Inventories and chemical groups	15		
	II.3	Accident sources during power operations			
	-	Gap release	18		
	-	In-vessel release	18		
	-	Vessel failure	19		
	-	Ex-vessel release	20		
	-	Air ingression release			
	-	Late in-vessel release			
	-	Iodine partitioning			
	II.4	Sources during shutdown accidents.			
	II.5	Chemical and physical forms of the released radionuclides			
	II.6	Annotated bibliography for sources	26		
III.	MIT	IGATION OF IN-VESSEL RELEASE	33		
	III.1	Coolant additions	33		
	III.2	Enhanced phoretic deposition.	34		
		Cooling of the vessel			
	III.4	Annotated bibliography on in-vessel retention processes	36		
IV.	MIT	IGATION OF EX-VESSEL RELEASE	37		
	IV.1	Introduction	37		
	IV.2	Pressure-driven debris expulsion	37		
	IV.3	Core debris interactions with coolant	38		
	IV.4	Core debris interactions with concrete	38		
	IV.5	Core retention devices	40		
	IV.6	Annotated bibliography on mitigation of ex-vessel releases	41		
V.	DEC	ONTAMINATION OF THE CONTAINMENT ATMOSPHERE	43		
	V.1	Introduction	43		
	V.2	Natural aerosol processes in the containment	43		
		2.1 Particle growth and gravitational sedimentation			
		2.2 Turbulent deposition	46		
		2.3 Phoretic deposition of particles	46		
	V.3	Engineered safety systems	46		
		3.1 Containment sprays	47		
		3.2 Steam suppression pools	47		
		3.3 Fan coolers			
		3.4 Effects of combinations of engineered safety systems			
	V.4	Annotated bibliography of aerosol removal from the containment atmosphere	50		

V1.	MIT	IGAT	FION OF GASEOUS IODINE	53
	VI.1	Intro	oduction	53
	VI.2	Iodii	ne chemical and transport behaviour within containment	54
		2.1	Aqueous phase reactions of iodine	
		2.2	Key parameters affecting net volatile iodine production	56
		2.3	Iodine behaviour in the gas phase	
	VI.3	Con	tainment iodine codes	
	VI.4	Rem	noval of gaseous iodine by filters	61
	VI.5	Sum	mary of Potential Mitigation methodologies	62
	VI.6		ident management as practised in OECD Member countries	
		6.1	Practises and applications adopted by the Member countries	64
		6.2	Spray systems	
		6.3	Containment sump additives	64
		6.4	Containment filters	
		6.5	Passive iodine removal	65
		6.6	Iodine mitigation measures implemented/planned	65
		6.7	Measurement of iodine in containment atmosphere and	
			controlled iodine release into the environment	65
		6.8	Other accident management measures to mitigate airborne iodine	65
		6.9	Conclusions	
	VI.7	Ann	otated bibliography of iodine chemistry in reactor accidents	72
VII.	MIT	IGAT	TION OF RELEASE FROM THE CONTAINMENT	77
			gation of At-power accidents	
			gation of release during shutdown accidents	
			otated bibliography on mitigation of release from containment	
	APP	ENDI	IX A	83
			n provided by Belgium, Canada, the Czech Republic,	03
			rance, Germany, Japan, the Republic of Korea, The Netherlands,	
			eden, Switzerland and the USA	
	Span	i, 5 W	den, ownzerrand and the obri	

I. INTRODUCTION

I.1 Accident management

Severe accident management is the use of existing and alternative resources, systems and actions to arrest or mitigate accidents that exceed the design basis of a nuclear power plant [In-1,2,3,4]. Most discussions of accident management are directed toward influencing the progression of accidents. The objectives of accident management in these usual discussions are to assure the reactor core is neutronically shutdown, cool the core, and maintain the core in a coolable configuration. There are, of course, substantial differences in how this might be accomplished in nuclear power plants of different types. There are, furthermore, significant national differences in the approaches to accident management. In America, for instance, accident management is a voluntary, industry undertaking that is mandated to involve minimal investment. On the other hand, in many European countries substantial capital investments are being made in accident management. Very often significant differences arise in the definition of actions that constitute accident management and actions that are part of the nuclear facility's established, emergency procedures.

A hallmark of accident management strategies is the consideration of diverse approaches to arrest or at least mitigate severe reactor accidents. A very direct way to mitigate the consequences of a nuclear power reactor accident is to attenuate the amount of radioactive material that is released into the environment even if the progression of the accident cannot be stopped. Accident management to attenuate the radioactive source term to the environment is the subject of this report. The attentions of this report, then, are upon very unlikely accidents that have progressed sufficiently that there has been or there might be some fuel damage and the release of significant amounts of radioactive material from a plant.

Accident management strategies to arrest or attenuate the releases of radioactive material from a reactor core or from a reactor containment are not developed in this report. Rather, the report delineates the technical bases for activities that might be included in such strategies. The report identifies actions that might affect the inventory of radioactive material that could escape the plant and provides a guide to the considerable body of literature pertinent to radionuclide behaviour under accident conditions. The annotated bibliographies presented here are by no means intended to be comprehensive. Instead the bibliographies should be considered mere introductions to the large and growing literature base on accident source terms and accident management. Readers are expected to resort to this literature base to obtain details that will be needed for the complete justification and development of accident management strategies.

Most of the discussions are presented here in the context of radionuclide behaviour during accidents at existing pressurised water reactors (PWRs) and boiling water reactors (BWRs). The basic principles in these discussions are applicable to all nuclear power plants. Readers may need to make some mental modifications of the specific details of the discussions to accommodate the unique features of other types of plants such as gas-cooled reactors, CANDU type reactors and RBMK reactors.

Recent probabilistic risk assessments [In-5-9] establish that there is room for accident management to influence the consequences of reactor accidents. These risk assessments show there to be orders of magnitude uncertainty in the radionuclide releases as a result of accidents at nuclear power plants. A very significant portion of this uncertainty comes from uncertainty in the details of radionuclide behavior that can be influenced by accident management measures. The accident management measures taken to affect the radionuclide source term are not necessarily distinct from those taken to arrest or

inhibit the progression of an accident. Indeed, it is argued here that radionuclides may be more sensitive to accident management measures than is core cooling. Insufficient credit may have been given in the past for the effects of core cooling accident management measures on both the release of radionuclides from reactor fuel and the behaviour of these released materials within the reactor coolant system and the reactor containment. On the other hand, accident management measures taken to cool the reactor fuel within the reactor vessel can actually enhance the release of radioactivity in some cases. Enhanced release usually occurs because vaporising coolant augments mass transport of radioactive vapours and aerosols out of the reactor coolant system.

Risk¹ has become the favoured metric for the evaluation of low probability but very high consequence events like severe reactor accidents. It is proving to be a metric of great value in the rational allocation of resources including allocation of accident management resources. The discipline inherent in quantitative risk assessment leads to a more comprehensive view of the needs and possible strategies of accident management than conventional, deterministic analyses that examine only individual components, systems and structures. The development of accident management strategies requires this more integrated view to avoid measures that are beneficial in one part of a facility or in one stage of an accident only to be surprised to find that these same measures exacerbate hazards in another part of the facility or in some later stage of an accident. Systematic risk evaluations provide the boundary conditions for accident management activities that might address situations as diverse as electrical power is not available (station blackout accidents), the containment is bypassed and no longer functions as a barrier to fission product releases (bypass accidents), safety systems and diagnostics are off-line for maintenance (accidents during shutdown operations), or support capabilities are not available (accidents initiated by earthquake).

Radionuclides are released to the containment as gases and as aerosol particles by a variety of processes during severe accidents. Modern, mechanistic analyses of these radionuclide releases and the subsequent behaviour of aerosols and vapours under reactor accident conditions strive to be realistic. This realistic approach contrasts with the deliberate attempt to be conservative (which may not have been successful) in the definition of radionuclide behaviour for the design of nuclear power plant safety systems. A discussion of the various radionuclide release processes during severe reactor accidents is presented in Chapter II. Of primary interest in these discussions of release is the potential magnitude of radionuclide release and the radionuclides of most concern. Factors that most affect radionuclide release but can also be affected by accident management measures are discussed.

The technical bases for possible accident management measures during the various stages of a severe accident are discussed in Chapters III to VII. These discussions follow the progression of severe accidents beginning with initial core degradation, progressing through core debris behaviour outside the reactor coolant system and concluding with radionuclide release from open or faulted containments. The attentions in these chapters are on factors that affect retention of radionuclides within a facility and accident management measures that can facilitate these retention processes.

^{1.} Risk is used here to mean the sum over the products of accident frequency and consequences. Risk is, then, distinct from just the frequency of accidents.

I.2 Annotated bibliography on accident management

In-1. **Accident Management Programmes in Nuclear Power Plants - A Guidebook**, Technical Reports Series No. 368, International Atomic Energy Agency, Vienna, Austria, 1994.

This report describes the steps to be taken in setting up an accident management program at a nuclear power plant. It focuses on the process rather than the specifics of the program since these specifics are likely to be unique to the power plant and its location.

In-2. W.E. Kastenberg, G. Apostolakis, V.K. Dhir, D. Okrent, M. Jae, H. Lim, T. Milici, H. Park, J. Swider, L. Xing, and D. Yu, A Framework for the Assessment of Severe Accident Management Strategies, NUREG/CR-6056, University of California, Los Angeles, CA, 1993.

This report discusses the evaluation of limited scope accident management programs of the type common in the USA. The report introduces the concept of the influence diagram for the evaluation of these programs. Such influence diagrams may have wider applicability even to more ambitious accident management programs. See also D.J. Hanson et al., **Developing and Assessing Accident Management Plans for Nuclear Power Plants**, NUREG/CR-6009, EGG-2682, Volumes 1 and 2, Idaho National Engineering Laboratory, Idaho Falls, ID, August 1992.

In-3. OECD Nuclear Energy Agency, **Proceedings of the Specialist Meeting on Severe Accident Management Implementation**, NEA/CSNI/R(95)5, June 1995.

This conference proceeding includes papers on accident management programs throughout the world. Conference papers can, of course, provide only overviews of the programs. References found in these papers often provide much greater detail.

In-4. D. Wach, "Variety of Tasks, Methods and Tools for Accident Management Support Progress Made in the AMS-Project", p. 377 in Reinforced concerted action on reactor safety FISA 95
 EU research on severe accidents, edited by G. Van Goethem, W. Balz, and E. Della Loggia, EU 16896 en, Directorate-General for Science, Research and Development, 1996.

This paper concentrates on the support available to operators during an accident. It does, however, provide some insight into the ambitious accident management efforts undertaken in many European countries.

In-5. Office of Nuclear Regulatory Research, Severe Accident Risks: An Assessment for Five U.S. Nuclear Power Plants, NUREG-1150, Volumes 1 and 2, U.S. Nuclear Regulatory Commission, Washington, D.C., June 1989.

This document provides an overview of the results of Level 3 probabilistic risk assessments for operational events including comprehensive uncertainty analyses for three types of PWRs and two types of BWRs. Among the results presented in the document are demonstrations of the uncertainties in the amounts of radioactive materials released from a plant in the risk dominant categories of accidents. Accident management measures can be designed to avoid the upper limits of the uncertainty ranges for these releases. It is unfortunate that similarly detailed probabilistic analyses of accidents during shutdown and low power operations are not available to support the design of accident management strategies.

In-6. Office of Nuclear Regulatory Research, **Individual Plant Examination Program: Perspectives on Reactor Safety and Plant Performance**, NUREG-1560 volume 1, Part 1, Draft Report for Comment, U.S. Nuclear Regulatory Commission, Washington, D.C., October 1996.

The results of 74 Level 1 probabilistic risk assessments for operational events at U.S. nuclear power plants are summarised in this report. The summary provides a good indication of the frequency dominant accidents in existing power plants and the ranges of predicted vulnerabilities to these accidents. There is an analogous effort now underway to address risk from events initiated by so-called external events such as earthquake, fire, and flooding. Such comprehensive studies of risk during shutdown and low power operations are not now planned.

In-7. L.M.C. Dutton, S.H.M. Jones, and J. Eyink, **Plant Assessments, Identification of Uncertainties in Source Term Analysis**, EUR 16502 en, European Commission, Luxembourg, 1995.

This report examines the severe accident sequences and radionuclide source terms at the Sizewell pressurised water reactor with a prestressed concrete containment, the Konvoi pressurized water reactor with a steel primary containment, the European Pressurised water Reactor (EPR) and a boiling water reactor with a Mark 2 containment.

In-8. Office of Nuclear Regulatory Research, **Individual Plant Examination Program: Perspectives on Reactor Safety and Plant Performance**, NUREG-1560, Volume 1, Part 1, U.S. Nuclear Regulatory Commission, Washington, D.C., October 1996.

This report summarises results of the Individual Plant Examinations for 74 nuclear power plant sites in the USA. The Individual Plant Examinations all involved Level 1 probabilistic risk assessments. Some also included level 2 analyses. The report describes the ranges of core damage frequencies in terms of the plant and containment types. The report provides a good indication of the types of frequency dominant accidents that might have to be addressed in the development of accident management strategies.

In-9 "NPP Temelín Safety Upgrading Programme and PSA Insights", IAEA Technical Meeting on Physical and Functional Separation of Safety Systems for WWER-1000 Reactors, International Atomic Energy Agency, Vienna, Austria, August 25-29, 1997 and for the Dukovany unit in W.J. Puglia et al., Analysis of Core Damage Frequency: Nuclear Power Plant Dukovany, VVER-440/213 Unit 1 - Internal Events, SAIC/94-6653, Science Applications International, December 1994. Procedures for Analysis of Accidents in Shutdown Modes for WWER Nuclear Power Plants, IAEA-EBP-WWER-09, International Atomic Energy Agency, Vienna, Austria, 1997.

These documents provide information on severe reactor accidents at two VVER type plants.

II. THE SOURCES

II.1 Introduction

An understanding of the physical and chemical natures as well as the magnitudes of radionuclide releases to reactor coolant systems and reactor containments is helpful in the design of strategies to mitigate the consequences of radionuclide release from nuclear power plants. The understanding of the sources of radionuclides under reactor accident conditions has undergone revolutionary changes over the last two decades. Primitive assessments that were hoped to be bounding [S-1] and were used to formulate the design of safety systems for most of today's nuclear power plants have been replaced by more detailed, mechanistic models of radionuclide behavior [S-2a,b]. Research into the details of radionuclide release is needed [S-2b] and is continuing [S-3]. Consequently, models used to predict the behavior of radionuclides under accident conditions are being improved regularly². In subsequent sections of this report, it will become apparent that the improved understanding of the sources of radionuclides is making possible the design of advanced strategies for more effective mitigation of accident consequences.

Early reactor safety assessments [S-1] hypothesised that severe accidents would entail the prompt release of a significant fraction of a bounding radionuclide (typically iodine) to the reactor containment. Safety systems were designed, then, for massive, immediate response to this release. Now, it is understood that radionuclide releases will take place by multiple processes over protracted periods and will involve many different radionuclides in different chemical and physical forms. Mitigation methods will have to operate for long periods and may have to change as the sources of radionuclides vary. The inventories of radionuclides available for release from reactor fuel under accident conditions and the processes that lead to releases of these radionuclides are discussed in the next subsections of this report.

II.2 Inventories and chemical groups

There are, of course, many radioactive isotopes available in irradiated reactor fuel. It is useful, then, to have some understanding of which of these radionuclides have the greatest importance to risk. Results of a particular, relative assessment of the radiological significance of various radioactive elements are shown in table III-1 [S-4]. The relative radiological effects of the release of 10% of the initial core inventory of several radionuclides are shown in this table. The selection of 10% release of each radionuclide inventory was arbitrary except that in each case it is sufficient to produce nonstochastic health effects beyond the site boundary of typical nuclear power plants. Early health effects of the releases have been normalised to the release of 10% of the reactor core inventory of iodine. Latent health effects have been normalised to the effects of releasing 10% of the initial core inventory of cesium. Though attentions often focus on the more volatile radionuclides such as cesium and iodine, it is apparent from the comparison in Table II-1 that other radionuclides can have as much or more impact should they escape a nuclear power plant. On the other hand, noble gases and some of the other radionuclides have relatively small health effects. Some conclusions about these other radionuclides might well change if land contamination effects were included in the comparison. Dutton et al. [S-2b] provide another way to assess the relative importances of radionuclides by examining their contributions to various measures of dose (thyroid, bone marrow, collective effective dose, etc.).

^{2.} Because models are often updated or replaced, it is difficult to compare risk assessments of different vintage. Models of varying technical sophistication have been used to predict radionuclide release and behavior.

Table II-1. Relative Radiological Importances of Radioactive Elements[S-4]

Element	Normalised 4 Hour Bone Marrow Dose	Normalised 24 Hour Bone Marrow Dose	Normalised Lung Dose	Normalised Total Latent Cancers
Kr	0.2	0.1	0.04	0.001
Xe	0.02	0.01	0.005	1×10^{-4}
I	1.0	1.0	1.0	0.1
Cs	0.15	0.14	0.09	1.0
Rb	$2x10^{-4}$	$2x10^{-4}$	$2x10^{-4}$	$1x10^{-5}$
Te	0.8	0.8	0.6	0.1
Sb	0.06	0.06	0.1	0.004
Sr	1.0	0.7	0.9	0.7
Ba	0.6	0.5	0.6	0.2
Ru	0.3	0.3	3.0	1.0
Rh	0.01	0.01	0.08	0.004
Mo	0.1	0.1	0.7	0.06
Tc	0.02	0.03	0.03	0.06
La	1.1	1.2	1.6	0.08
Y	0.07	0.07	3.5	0.4
Nd	0.03	0.03	0.3	0.03
Pr	0.004	0.003	0.8	0.08
Ce	0.1	0.2	8.0	2.0
Zr	1.0	1.0	2.0	0.7
Pu	0.004	0.003	1.4	3.0
Np	1.6	1.4	5.0	0.4
Cm	0.6	0.4	5.0	1.1

It is important to remember that, for the purposes of accident management, it is the product of an element's radiological significance and its release fraction that determine its risk significance. To focus accident management activities on the most risk significant elements, then, it is necessary to have some understanding of the potential releases of the elements under accident conditions. Each of the radioactive elements in irradiated reactor fuel has its own, unique chemistry that dictates its release behaviour. These chemistries are, however, sufficiently similar that it is possible to group radioactive elements into categories to simplify discussions. Many different groupings have been considered. The grouping often adopted [S-2] is:

Group Name	Radioactive Elements
Noble gases	Xe, Kr
Halogens	I, Br
Alkali Metals	Cs, Rb
Alkaline Earths	Sr, Ba
Chalcogens	Te, Se, Sb, As
Refractory Metals	Ru, Mo, Pd, Tc, Rh
Lanthanides	La, Y, Sm, Gd, Nd, Pr, Eu
Actinides	Ce, Pu, U, Th, Zr, Cm

Note that the elements within a group do not necessarily conform to the usual chemical constituents of the group of the same name. The release behaviors of these elements have usually been found sufficiently similar to be included in the group. Sometimes alternative groupings of the elements are developed to better reflect the evolving understanding of chemistry under accident conditions.

An even simpler categorisation of radionuclides has also been used. The simplest categorisation is to treat the radionuclides that could be released from a nuclear power plant as a mixture of noble gases, gaseous iodine, cesium particulate and other particulate. Little can be done by way of accident management to attenuate the release of noble gases³ except to reduce the leakage from the reactor containment (See Chapter VII). Gaseous iodine release can be attenuated by trapping in water, adsorption on surfaces, chemical transformations and filtration (See Chapter VI). Most of the source term measures of accident management are focused on reducing the amounts of radioactive particulate that can be released from the reactor and discussions of the physical and chemical bases of these measures make up much of the rest of this document.

^{3.} There have been studies of methods to trap noble gases. In the main, absorption methods using zeolites or charcoal have been examined. Though the noble gases will absorb on these materials, it has proved challenging to engineer systems that will operate well under the range of conditions expected to arise in severe accidents.

Understanding of the sources of radionuclides in reactor accidents is best for accidents initiated during power operations by either internal or external events. Less is known about sources during accidents initiated during shutdown and low power operations. The understanding of sources during power operation accidents is discussed in the next subsection. To a large extent these discussions are expected to have generic applicability. Peculiarities of sources during accidents initiated during shutdown operations are discussed in a subsequent subsection.

II.3 Accident sources during power operations

The first release of radionuclides to the reactor containment comes from this discharge of contaminated coolant from the reactor coolant system. This discharge may be through a break in the reactor coolant system or through pressure relief valves. Contamination of the coolant comes from activation and from the leakage of radionuclides from failed fuel pins. The level of contamination is quite small since coolant contamination and the incidences of rod failure during power plant operation are usually tightly controlled by regulation. Certainly in comparison to the releases of radionuclides that occur in subsequent stages of reactor accidents, the releases of radionuclides associated with coolant discharge are minuscule. The releases associated with coolant discharge are often neglected in the analyses of reactor accidents. They may have more significance in the design of accident management strategies. Certainly, the discharge releases and the activation of radiation alarms in the containment provided the first definitive evidence to the operators of serious events during the accident at Three Mile Island [S-5].

Radionuclide releases following coolant discharge from the reactor coolant system are usually categorized according to the stage of accident progression. These stages of accident progression and the associated releases of radioactivity are as follows:

Gap release

Once an accident is initiated, coolant levels drop below the top of the active fuel. The cladding on the fuel ruptures and the gap inventory of radionuclides is released to the flow through the reactor coolant system. The gap release consists primarily of noble gases and the more volatile radionuclides such as cesium, iodine, and perhaps tellurium. Some finely fragmented fuel particles may also be released. Gap inventories have been the subject of debate for the last 20 years [S-6]. Some of the best experimental studies of gap release have been reported by Malinauskus and coworkers [S-7]. These studies and available models of gap release may, however, become obsolete as reactors use fuel to higher levels of burnup and different linear heating rates than have been anticipated. High fuel burnups produce microstructure adjacent to the fuel clad that appear conducive to much higher gap inventories. Models of fission gas release such as FREEDOM and the GRASS code series may have to be modified to predict gap inventories accurately in the first, serious, stage of radionuclide release.

In-vessel release

Decay heating will raise the temperature of the fuel at rates that are usually less than 1 K/s. Eventually, a temperature is reached at which the exoergic reaction of steam with zirconium alloy cladding is limited only by the mass transport of steam to the clad. Fuel then experiences a temperature excursion at rates of 20 K/s or more. Diffusion of radionuclides through solid and even liquid fuel produces a pronounced increase in the radionuclide release at these elevated temperatures. As discussed further in Chapter III, only a fraction of the radionuclides released from the fuel successfully negotiates

passage through the reactor coolant system and into the containment. The rest is deposited within the coolant system. This may, however, be only a temporary holdup of released radionuclides (See Late In-vessel Release, below).

Substantial quantities of nonradioactive material may be vaporized from the reactor core during this stage of an accident. In particular, control rod materials and burnable poisons may be vaporised. Constituents of structural materials such as steel and clad alloying agents as well as uranium oxides may be vaporised along with radionuclides. These nonradioactive materials add to the mass of condensable effluents from the core region and can affect the behavior of radionuclides both in the reactor coolant system and in the containment (See Chapter V).

Core degradation during this stage of an accident has been the subject of many in-pile (PBF; PHEBUS) and out-of-pile (CORA; HI; VI) experimental studies. There is now some confidence that, without accident management efforts and prior to the loss of core geometry, understanding of the degradation process is adequate for most purposes. This confidence is tempered somewhat by evidence that highly irradiated fuel may degrade differently than lower burnup (<40 Gwd/t) fuels studied to date⁴. The degradation studies often, also, provided data on releases associated with the core degradation. The ongoing PHEBUS-FP program is providing a dedicated examination of releases and radionuclide behavior during this stage of a reactor accident [S-8]. Mechanistic models of radionuclide release and transport during this phase of an accident such as VICTORIA, SOPHAEROS, and ART [S-9] are being developed.

Although the in-vessel stage of a reactor accident has occupied a significant amount of attention in the development of accident management strategies, there remains uncertainty in the understanding of the degradation process if the supply of coolant is inadequate to quench the overheated fuel. Limited amounts of coolant may exacerbate the metal-water reaction responsible for rapid heating of the fuel by augmenting the supply of steam. The ongoing QUENCH program is investigating the reflood phenomena. There is also not a clear understanding of whether the collapse of brittle fuel, as was observed during the accident at Three Mile Island, will interfere in the ability to cool the degraded core even if sources of coolant become available [S-10]. The effects of early melting of the control rods (both silver-indium-cadmium and steel-clad boron carbide control rods and blades) during the degradation process is also an area of some uncertainty.

Vessel failure

If the accident cannot be arrested, core debris will accumulate in the lower plenum of the reactor vessel. It is usually estimated that something less than 50% of the core debris will collapse into this plenum which in many accidents will still contain water. Violent fuel coolant interactions could occur when hot or even molten core debris falls into the lower plenum. The structural consequences of such interactions are the topic of ongoing debate [S-11]. Assuredly, fuel-coolant interactions could produce dramatic increases in the flow through the reactor coolant system. These abrupt increases in flow will purge the reactor coolant system of suspended radionuclide vapours and particles. Flows might be

^{4.} The region of the core undergoing degradation will contain, typically, both high and lower burnup fuel. Releases of radionuclides are averages over this fuel mix which attenuates any errors in the modeling of the high burnup fuel.

sufficient to resuspend radionuclides deposited in the reactor coolant system and carry these radionuclides into the containment. There have been [S-12] and are ongoing studies of the resuspension of deposited radionuclides by sudden increases in flow [S-13].

There is thought now that especially for reactors with fuel having a low power density it may be possible to arrest accidents by simultaneously providing coolant to the core and by cooling the outside of the reactor vessel [S-14]. The radionuclide release consequences of prolonged retention of core debris in the reactor vessel have not received much attention.

If core debris in the lower plenum cannot be quenched quickly, it will eventually penetrate the reactor vessel. If the reactor coolant system is pressurised to levels above the pressurisation of the reactor containment, a pressure-driven expulsion of core debris will accompany vessel rupture. Studies of pressurised expulsion of simulant materials from a vessel suggest that 1-9% of the expelled material could be converted into aerosol by a combination of mechanical and vaporisation processes [S-15]. Interest in this source of radionuclides to the containment atmosphere has waned as procedures have been adopted to assure the reactor coolant system is depressurised during an accident.

Interaction of expelled core debris with water could inject radioactive aerosols into the containment atmosphere as a result of either steam explosions or just debris fragmentation and elutriation by rapidly generated steam. Earlier suggestions that this might be a mechanism for the extensive release of radioactive ruthenium from core debris [S-16] have been questioned by more recent analyses [S-17].

Ex-vessel release

Core debris interacting with structural concrete produces large amounts of gas by vaporising water that reacts to form hydrogen and decomposing whatever carbonates are in the concrete aggregate (some concretes have as little as 1% carbonate and others have as much as 36% carbonate) to produce carbon dioxide and carbon monoxide. These gases will sparge through the core debris and lead to the formation of large quantities of aerosol from both radioactive and nonradioactive materials. This release during core debris interactions with concrete has received quite a lot of study because it is expected to continue for long periods and because it is amenable to experimental study [S-18,19]. It is known that for much of the time aerosols are mostly(>95%) nonradioactive material. The most vigorous release of radionuclides occurs when metallic zirconium that was not oxidised during the in-vessel phases of the reactor accident is present in the core debris as it attacks concrete.

Air ingression release

Once the reactor vessel has been penetrated by core debris, it is possible for air from the reactor containment to circulate into the vessel and interact with both the residual fuel (perhaps more than 50% of the fuel inventory) and with radionuclides deposited in the reactor coolant system. Even containment atmospheres that have been "inerted" to suppress hydrogen combustion contain about 5% air, which is sufficient to be of concern for air ingression scenarios. It has been suggested that air interactions with residual fuel could lead to the very extensive release of ruthenium and molybdenum from this fuel - far more release than is possible in the steam/hydrogen environment during the earlier core degradation stage of the accident [S-20, 21]. It is also possible that air will convert deposited iodides in the reactor coolant system into elemental iodine gas. These possibilities are being investigated as part of the current PHEBUS-FP program.

Late in-vessel release

Decay heating of deposits in the reactor coolant system can produce temperatures in these deposits sufficient to cause extensive revaporisation of volatile radionuclides such as cesium, iodine and perhaps tellurium and antimony. This revaporisation process can lead to a protracted, low intensity release of radionuclides to the containment. Some analyses have suggested that significant release by this mechanism could continue for periods of up to 50 hours [S-22]. Thus, revaporisation can maintain risk significant concentrations of radionuclides in the reactor containment for very long times. Because of this, revaporisation is viewed as a more significant source term than resuspension of deposited radionuclides. The revaporisation release is the subject of study both in work associated with the PHEBUS-FP program and in a Concerted Action program sponsored by the European Communities.

Iodine partitioning

As will be discussed at greater length in Chapter VI of this report, there are natural and engineered systems that drive most radionuclides released to an intact containment into water. Once in water, most of the radionuclides are permanently trapped unless core debris interactions with concrete penetrate the reactor basemat and provide a release pathway for the contaminated water. Iodine (and possibly tellurium and ruthenium) can behave differently. Hydrolysis and radiolysis processes can convert iodine dissolved in water into volatile elemental iodine (I₂) or volatile organic iodides (such as CH₃I). These volatile species can partition from the aqueous phase back into the containment atmosphere and thereby create a continuing source of radioactive material available for release in the event of containment failure. This continuing source to the containment atmosphere can develop even when the accident is otherwise arrested at an early stage. The continuing source can frustrate efforts to cleanse the atmosphere and recover from an accident.

There are, then, several potential sources of radioactive material to the containment atmosphere. The most important of these are thought to be gap release, in-vessel release, ex-vessel release, and late invessel release. A recent assessment of the magnitudes of these releases in terms of fractions of the initial core inventories of important radionuclides has been prepared [S-2]. Results of this assessment are shown in Tables II-2 and II-3. It is important to view the results shown in these tables as somewhat conservative examples of both the timing and the magnitude of radionuclide release. These examples are not prescriptions of accident sources to any particular reactor containment.

A source of fission products to the reactor containment that is not often considered is release from a spent fuel pool located within the containment when pool cooling and residual heat removal cannot be maintained. This configuration of the spent fuel pool within the containment is found in the VVER-1000 reactors. Overheated fuel in the spent fuel pool can undergo gap release, release of radionuclides during fuel degradation and release as a result of molten fuel interactions with concrete. Physical and chemical phenomena associated with these stages of release are likely to be quite similar to releases usually considered in safety analyses except the ambient atmosphere will be air rather than steam [S-25]. No detailed analyses of severe fuel degradation and radionuclide release during accidents involving fuel in a spent fuel pool like that found in VVER-1000 reactors have been published.

Finally, it should be remembered that releases of radionuclides from a plant may not be just airborne releases of gases and aerosols. Basemat penetration by core debris can lead to drainage of contaminated water into the ground. An accident management activity that might be considered is the interdiction of this contaminated water before it can reach other water bodies either on or below the surface.

II.4 Sources during shutdown accidents

The unmitigated progression of a reactor accident initiated during shutdown operations would be much like an accident initiated during power operations. Concerns over pressurised melt expulsion are less because during shutdown it is likely that the reactor coolant system is not pressurised and may even be open to the containment so that it does not become pressurised as the accident progresses. Steam explosions become of more concern because inhibitions to triggering are less at low pressure. The possibility of air intrusion into the reactor vessel during the earliest stages of core degradation is somewhat greater than in accidents during power operations. Mitigation of release of radionuclides to the containment by deposition in the reactor coolant system may be less effective depending on how open the reactor coolant system is.

It is, however, very likely that accidents initiated during shutdown will be promptly detected and arrested. There will be, after all, a large workforce operating around the reactor during shutdown. The system will be open and at low pressure, so water injection from even nontraditional sources can be used to arrest the accident. Arresting the accident will, however, involve exposure of brittle clad and fuel to quenching which could result in fragmentation of the core. This, in turn, will expose bare fuel to water. Fission products can leach from the fuel. Of particular concern is the leaching of fission products such as iodine that can undergo radiolysis and hydrolysis processes to produce volatile forms that partition into the containment atmosphere. Under shutdown conditions this is a particularly serious event since the containment is likely to be open to the environment and the partitioned radionuclides can readily escape. The continuing source of iodine (and, perhaps, some other radionuclides) could hinder further efforts to arrest the release.

It should also be noted that releases of radioactivity from spent fuel pools can also occur during shutdown conditions if decay heat removal is not maintained.

II.5 Chemical and physical forms of the released radionuclides

Details of the chemical and physical form of the released radionuclides are receiving a great deal of attention. These details need to be known to predict the retention in the reactor coolant system, the revaporization source and the resuspension source. Information needs for the development of accident management strategies are probably less demanding. For many purposes, it can be sufficient to class radionuclides as:

- noble gases that really defy mitigation, but pose less threat;
- Iodine that can be present as either a gaseous or as an aerosol species; and
- aerosols.

Aerosol behaviour in the containment is largely dictated by physical properties including:

- size distribution of the particles;
- particle shapes, drag and mobility;
- particle density;
- particle thermal conductivity; and
- electrostatic charging of the particles.

The only major chemical issues concerning aerosol particles within the reactor containment are questions of hygroscopicity and the revaporisation of aerosol constituents exposed to hydrogen burns. Unfortunately, beyond some relatively imprecise information about particle size there are not good databases on the physical properties of aerosol particles likely to be produced in reactor accidents and no validated models of these properties. A more troublesome issue is the question of electrostatic charging of the aerosols because of their innate radioactivity and because of the intense ambient radiation field. Preferential charging of aerosol particles could make it necessary to add Coulombic terms in the equations that describe aerosol growth and agglomeration [S-23]. The electrostatic charging of aerosol particles, if it occurs to a significant extent, would be a unique feature of aerosols in reactor accidents. Certainly, it would be a feature of the aerosol that would be difficult to reproduce accurately in conventional aerosol test facilities that have been used to validate models now used for reactor accident analyses. It may be that the very prototypic tests now planned for the PHEBUS-FP program will cast light on the importance of electrostatic charging on aerosol behaviour in reactor containments.

There are, then, still uncertainties in the predictions of aerosol behaviour in reactor accidents. Consequently, accident management efforts must be developed to deal with aerosols having a broad range of properties. There are similarly large uncertainties concerning the chemical forms adopted by the released radionuclides. Though research continues in this area, accident management strategies should be developed to confront a broad range of possible chemical forms of the radionuclides and non-radioactive materials released from the reactor fuel.

Table II-2. Accident sources to the containment for pressurised water reactors [S-2]

	Gap Release	In-vessel Release	Ex-vessel Release	Late In-vessel Release
Duration(seconds)	1800	4680	7200	36000
Release Fractions of Noble Gases	0.05	0.95	-	-
Halogens	0.05	0.35	0.25	0.10
Alkali Metals	0.05	0.25	0.35	0.10
Alkaline Earths	-	0.02	0.10	-
Chalcogens	-	0.05	0.25	0.005
Refractory Metals	-	0.0025	0.0025	-
Lanthanides	-	0.0002	0.005	-
Actinides	-	0.0005	0.005	-

Table II-3. Accident sources to the containment for boiling water reactors [S-2]

	Gap Release	In-vessel Release	Ex-vessel Release	Late In-vessel Release
Duration(seconds)	1800	5400	10800	36000
Release Fractions of Noble Gases	0.05	0.95	-	-
Halogens	0.05	0.25	0.30	0.01
Alkali Metals	0.05	0.20	0.35	0.01
Alkaline Earths	-	0.02	0.10	-
Chalcogens	-	0.05	0.25	0.005
Refractory Metals	-	0.0025	0.0025	-
Lanthanides	-	0.0002	0.005	-
Actinides	-	0.0005	0.005	-

II.6 Annotated bibliography for sources

S-1. J.J. DiNunno *et al.*, Calculation of Distance Factors for Power and Test Reactor Sites, TID-14844, U.S. Atomic Energy Commission, Washington, D.C., 1962.

This document describes the treatment of reactor accident source terms used in the past for both the design and safety evaluation of many existing power plants. The document, which is now only of historical significance, envisages the important radionuclide releases to be the noble gases, iodine in gaseous form, and 1% of the initial core inventory of other radionuclides in particulate form.

S-2a. L. Soffer, S.B. Burson, C.M. Ferrell, R.Y. Lee, and J.N. Ridgely, **Accident Source Terms for Light-Water Nuclear Power Plants**, NUREG-1465, U.S. Nuclear Regulatory Commission, Washington, D.C. February 1995.

This document describes a representative radionuclide source term to the containments of reactors for use in the traditional design basis accident analysis. The source term is based on the more mechanistic studies of radionuclide behaviour conducted since the accident at Three Mile Island. Although the source is representative of the magnitude and timing of severe accident source terms, it is not intended to represent any specific severe accident sequence nor is it intended to be a bounding source term.

S-2b. L.M.C. Dutton, S.H.M. Jones, and J. Eyink, **Plant Assessments, Identification of Uncertainties in Source Term Analysis**, EUR 16502 en, European Commission, Luxembourg, 1995.

This report examines the severe accident sequences and radionuclide source terms at the Sizewell pressurised water reactor with a prestressed concrete containment, the Konvoi pressurized water reactor with a steel primary containment, the European Pressurised water Reactor (EPR) and a boiling water reactor with a Mark 2 containment. The report concludes that the key accident sequences for European plant designs are transient events and small loss-of-coolant accidents, loss of cooling during shutdown, and containment bypass sequences. The most important chemical and transport phenomena are found to be revaporisation of volatile radionuclides from the reactor coolant system, iodine chemistry, and release paths through the plant. Additional research is recommended on release of fission products from the fuel, release of fission products from the reactor coolant system, chemistry of iodine, and transport of radionuclide through plants.

S-3. A.L. Wright *et al.*, **Primary System Fission Product Release and Transport - A State-of-the-Art Report to the Committee on the Safety of Nuclear Installations**, NUREG/CR-6193, NEA/CSNI/R(94)2, ORNL/TM-12681, Oak Ridge National Laboratory, Oak Ridge, TN, June 1994.

This report provides a comprehensive review of the current understanding of core degradation and fission product release and transport during severe reactor accidents. The report identifies areas of understanding and remaining uncertainties in both the experimental and the analytic areas. The new VEGA program to study release of non-volatile and short-lived fission products from fuels at temperatures up to 3000 °C and pressures up to 1.0 MPa is described in A. Hidaka, T. Nakamura, Y. Harada, and J. Sugimoto, "Status of VEGA Fission Product Release Experiment", **Proceedings of the Workshop on Severe Accidents (SARJ-97)**, JAERI-Conf 98-009, Japan Atomic Research Institute, 1998.

S-4. D.J. Alpert, D.I. Chanin, and L.T. Ritchie, **Relative Importance of Individual Elements to Reactor Accident Consequences Assuming Equal Release Fractions**, NUREG/CR-4467, SAND85-2575, Sandia National Laboratories, Albuquerque, NM, March 1988.

This report shows a basis for judging the relative importances of the many radionuclides. See also S-2b.

S-5. J.M. Broughton, P. Kuan, D.A. Petti, and E.L. Tolman, "A Scenario of the Three Mile Island Unit 2 Accident", **Nuclear Technology 87** (1989) 34.

A readable account of the accident and findings of the post-event dissection of the reactor coolant system. The article shows why there is some confidence that accident management actions can be expected to delay, mitigate or arrest accident progression and the release of radionuclides.

S-6. R.L. Ritzman *et al.*, "Release of Radioactivity in Reactor Accidents", Appendix VII, **Reactor Safety Study**, NUREG 75/014, WASH 1400, U.S. Nuclear Regulatory Commission, Washington, D.C., October 1975.

Despite the relative age of this report, it provides a good account of the current understanding of gap release during reactor accidents. Much of the information in this account becomes of decreasing utility as burnups of reactor fuel go beyond about 30 GWd/t.

S-7. R.A. Lorenz, J.L. Collins, A.P. Malinauskus, Nuclear Technology, 46(1979)404.

Experimental studies of gap releases are reported. The correlation of the results presented in this paper is widely used to estimate gap releases of radionuclides in reactor accident analyses. The reliability of the correlations of experimental data becomes questionable as the burnups and linear power ratings of fuels go beyond the underlying data base.

S-8. P. von der Hardt and A. Tattegrain, "The PHEBUS Fission Product Project", **J. Nuclear Materials**, *188* (1992) 115.

This paper provides an overview of the monumental experimental program to provide data to validate models of radionuclide behaviour under accident conditions. There is an enormous volume of literature concerning this ongoing program.

- S-9a. T.J. Heames, *et al.*, **VICTORIA: A Mechanistic Model of Radionuclide Behavior in the Reactor Coolant System Under Severe Accident Conditions**, NUREG/CR-5545, SAND90-0756, Rev. 1, Sandia National Laboratories, Albuquerque, NM, December 1992.
- S-9b. M. Missirlian and G. Lajtha, **SOPHAEROS Code V1.3 Theoretical Manual**, Département de Recherches en Sécurite, Cadarache, France, September 1, 1997
- S-9c. M. Igarashi, *et al.*, "Deposition of Cesium Iodide in Horizontal Straight Pipes Under Severe Accident Conditions", **5th International Conference on Nuclear Engineering**, Nice, France, May 26-30, 1997.

The VICTORIA, SOPHAEROS, and ART models are representative of the modern mechanistic models for the prediction of fission product release and transport to the reactor containment that have replaced the more *ad hoc* and bounding representations found in the design bases and safety analyses of most existing reactors.

S-10. P. Kuan, D.J. Hanson, D.J. Pafford, K.S. Quick, and R.J. Witt, **Implications for Accident Management of Adding Water to Degrading Reactor Cores**, NUREG/CR-6158, EGG-2644, Idaho National Engineering Laboratory, Idaho Falls, ID, February 1994.

This report discusses the positive and negative features of coolant addition as a means to arrest or mitigate a core degradation accident. It focuses on the accident progression aspects of accident mitigation and does not address some of the more important consequences of coolant additions on fission product release and behaviour. There does not seem to be in the literature a corresponding analysis of the source term issues associated with coolant additions.

S-11. N. Yamano, Y. Maruyama, K. Moriyama, and J. Sugimoto, Technical Note on Ex-Vessel Core Melt Debris Coolability and Steam Explosions, NEA/CSNI/R(96)24, Committee on the Safety of Nuclear Installations, OECD Nuclear Energy Agency, Issy-les-Moulineaux, France, December 1996.

This technical note provides a good introduction to the literature of the thermal and mechanical processes associated with high temperature melt interactions with water. For further information on such interactions see also **Proceedings of the OECD/CSNI Specialists' Meeting on Fuel-Coolant Interactions**, M. Akiyama, N. Yamano, and J. Sugimoto, editors, NEA/CSNI/R(97)26, May 19-21, 1997, Tokai-Mura, Japan. There has been little study of the implications of these interactions on the accident source term.

S-12. NEA Group of Experts, **Short Overview on the Definitions and Significance of the Late Phase Fission Product Aerosol/Vapour Source**, NEA/CSNI/R(94)30, Committee on the Safety of Nuclear Installations, OECD Nuclear Energy Agency, Issy-les-Moulineaux, France, September 1994.

This paper defines terms such as "resuspension", "revaporisation", "reentrainment", and "revolatilisation" and the role of the phenomena described by these terms in the late release of radionuclides from the reactor coolant system and reactor containment. The paper also has a bibliography of research done on this source term.

S-13. M. Eusebi, *et al.*, "Preparatory Calculations for a New Experimental Program on Dry Aerosol Resuspension Mechanisms (STORM Project)", **Proceedings of the European Aerosol Conference**, Oxford, United Kingdom, 1992.

This paper provides an introduction to an ongoing experimental program to obtain data for the validation of models of aerosol resuspension.

S-14. R.E. Henry, *et al.*, "Cooling of Core Debris within the Reactor Vessel Lower Head", **Nuclear Technology**, *101*(1993)385.

This paper provides an overview of the strategy for using external cooling of the reactor vessel to arrest the progression of an accident. More technical details can be found in the **Proceedings of the OECD/CSNI Workshop on In-Vessel Core Debris Retention and Coolability**, NEA/CSNI/R(98)21, Garching, Germany March 3-6, 1998. The focus in all of these sources is on accident progression. Very little attention has been given to the source term implications of the accident management strategy.

S-15. J.E. Brockmann, "Ex-Vessel Releases: Aerosol Source Terms in Reactor Accidents", **Progress in Nuclear Energy 19**(1987)7.

Though this paper is becoming somewhat dated, it still provides a good account about what is known concerning radionuclide release and aerosol formation during the ex-vessel stages of reactor accidents including steam explosions, pressure-driven melt expulsion and core debris concrete interactions. A good understanding of the thermal and mechanical aspects of the pressure-driven melt expulsion process and direct containment heating of dispersed core debris can be obtained from Fauske & Associates, Inc. And Sandia National Laboratories, **State-of-the-Art Report (SOAR) on HPME and DCH**, NEA/CSNI/R(96)25, OCDE/GD(96)194, Committee on the Safety of Nuclear Installations, OECD Nuclear Energy Agency, Issy-les-Moulineaux, France, December 1996.

S-16. R.L. Ritzman *et al.*, "Release of Radioactivity in Reactor Accidents" Appendix VII, **Reactor Safety Study**, NUREG 75/014, WASH 1400, U.S. Nuclear Regulatory Commission, Washington, D.C., October 1975.

The original analysis of possible releases associated with steam explosions is presented in this paper. Releases of this type have largely been discounted in most risk assessments. There have been no experimental studies to confirm or to refute the analyses.

S-17. D.A. Powers, "A re-examination of the steam explosion source term during severe accidents" p.391, **Source Term Evaluation for Accident Conditions**, International Atomic Energy Agency, Vienna Austria, 1986.

This re-examination of the analyses discussed in S-16 concludes that the source term from fuel-coolant interactions is likely to be small.

- S-18a. Proceedings of the OECD(NEA)CSNI Specialists' Meeting on Molten Core Debris-Concrete Interactions, KfK-5108, NEA/CSNI/R(92)10, Kernforschungszentrum Karlsruhe, Karlsruhe, Germany, 1992.
- S-18b. Proceedings of the Committee on the Safety of Nuclear Installations (CSNI) Specialists' Meeting on Core Debris-Concrete Interactions, EPRI NP-5054-SR, Electric Power Research Institute, Palo Alto, CA, February 1987.

The proceedings of these meetings provide a good indication of the range of experimental and analytical activities devoted to the study of core debris interactions with concrete. Most of the work has dealt with concrete ablation and combustible gas generation. Relatively less work has gone into the study of radionuclide release during core debris interactions with concrete.

S-19. D.A. Powers, J.E. Brockmann, A.W. Shiver, VANESA: A Mechanistic Model of Radionuclide Release and Aerosol Generation During Core Debris Interactions with Concrete, NUREG/CR-4308, SAND85-1307, Sandia National Laboratories, Albuquerque, NM, 1985.

This report, though dated, provides a thorough discussion of the physical phenomena that affect fission product release and aerosol formation during the interactions of core debris with concrete.

S-20. F.C. Iglesias, C.E.L. Hunt, and F. Garisto, "Measured Release Kinetics of Ruthenium from Uranium Oxides in Air", and C.E.L. Hunt, F.C. Iglesias, and D.S. Cox, "Measured Release Kinetics of Iodine and Cesium from UO₂ at High Temperatures Under Accident Conditions", in **Proceeding of the International Seminar on Fission Product Transport Processes During Reactor Accidents**, J.T. Rodgers, editor, Hemisphere Publishing Co., 1990.

These experimental studies illustrate the important consequences of air ingression into the reactor vessel when hot fuel residues are present. Ruthenium releases as well as the releases of some other radionuclides are greatly accentuated. The proceedings of the seminar provide also a good digest of much of the world-wide research on severe reactor accident source terms at the time.

S-21. D.A. Powers, L.N. Kmetyk, and R.C. Schmidt, **A Review of the Technical Issues of Air Ingression During Severe Reactor Accidents**, NUREG/CR-6218, SAND94-0731, Sandia National Laboratories, Albuquerque, NM, December 1994.

This report discusses analyses of the source term consequences of air ingression into the reactor coolant systems during severe accidents.

S-22. Industry Degraded Core Rulemaking Project, **Peach Bottom Atomic Power Station, IDCOR Task 23.1, Integrated Containment Analysis**, Atomic Industrial Forum, Bethesda, Maryland, 1984.

This document presents analyses of accidents in a Mark 1 BWR that show revaporisation releases of radionuclides deposited in the reactor coolant system and in the drywell can continue at significant rates for up to 50 hours.

S-23. C.F. Clement and R.G. Harrison, "The charging of radioactive aerosol", J. Aerosol Science, 23(1992)481.

This paper provides an introduction to the magnitude of electrostatic charging of aerosol particles during reactor accidents. The charge distribution varies from the Boltzmann distribution primarily because of differences in the mobilities of atmospheric ions produced by the radiation field in containment during accidents.

- S-24a. M. Kajimoto *et al.*, "Analysis of Aerosol Behavior in Containment Overpressure Failure Scenarios in BWR Mrk-II Plant", **NEA/CEC/CEA Workshop on Aerosol Behavior and Thermal Hydraulics in Containment**, CSNI Report 176, Paris, 1990.
- S-24b. M. Kajimoto *et al.*, "Analysis of Source Term Uncertainty Issues for LWRs", **Proceedings of the International Conference on Probabilistic Safety Assessment**, PSAM-II, San Diego, CA, 1994.

These papers provide the calculated results of source terms for a large number of accident sequences and illustrate the dependencies of the severe accident source terms on the accident sequence.

S-25. A. S. Benjamin, D. J. McCloskey, D. A. Powers, and S. A. Dupree, **Spent Fuel Heatup Following Loss of Water During Storage**, SAND77-1371, Sandia Laboratories, Albuquerque, NM, March 1979.

This report discusses some of the phenomenological issues that arise when cooling to the spent fuel pool is lost. Studies of radionuclide releases have not been reported.

III. MITIGATION OF IN-VESSEL RELEASE

III.1 Coolant additions

Most discussions of accident management focus on arresting the reactor accident while the core is within the reactor vessel. Indeed, many accident management strategies such as manual depressurisation and « feed and bleed » are attempts to prevent any significant degradation of the reactor core. The confidence that this can be done in the face of multiple systems failures springs, of course, from the successful termination of the Three Mile Island accident before core debris was able to penetrate the reactor vessel, even though preplanned accident management strategies and emergency operating procedures were not in place.

Strategies aimed at preventing or mitigating core damage all focus on getting coolant to the fuel to reduce the fuel temperature. To the extent that these measures do cool the fuel or, at least slow the temperature rise, they also mitigate radionuclide release that is so very dependent on temperature.

Addition of coolant has, however, a profound effect on mitigation of radionuclide release even if coolant additions are insufficient to cool the fuel. Indeed, analyses have suggested that coolant additions during critical stages of the in-vessel progression of an accident may accelerate core degradation. Still, these coolant additions may limit the amounts of radioactive material released from the fuel that passes through the reactor coolant system and reaches the containment. This mitigation comes about by several processes:

- coolant droplets sweep out radionuclide vapors and aerosols in the reactor coolant system;
- flooded release pathways decontaminate gases laden with radioactive vapours and aerosols;
 and
- cooled structural surfaces in the reactor coolant system act as deposition surfaces for radioactive aerosol particles.

Sweep out of aerosols and vapours by coolant droplets is entirely analogous to spray removal of aerosol particles from the containment atmosphere. The analogy is particularly strong when coolant additions are made through core sprays. The decontamination by spray droplets can be quite efficient, so coolant addition via sprays is an attractive accident management strategy. The physical phenomena and limitations of spray removal of particles and vapours are discussed at greater length in Chapter V.

Flooded release pathways created by coolant addition were responsible for the relatively modest radionuclide releases to the containment during the Three Mile Island accident. Gas flows through flooded pathways are broken into bubbles. Radioactive vapours and aerosols are removed from the bubbles by processes that are quite like those that occur in steam suppression pools. These removal processes are also discussed in Chapter V.

On the other hand, coolant additions insufficient to quench the degrading reactor fuel may enhance the release of radioactivity from the reactor coolant system. Enhanced releases can come about because the enhanced availability of steam for reaction with zirconium clad leads to higher fuel temperatures. Higher steam concentrations can make some radionuclides more volatile. Or, enhanced releases may be caused simply because higher steam production leads to more efficient mass transport through the reactor coolant system.

III.2 Enhanced phoretic deposition

Without either spray droplets or flooded pathways, substantial fractions of radionuclides released from the degrading reactor fuel can be retained within the reactor coolant system. Results of some example calculation for radionuclide retention in the reactor coolant systems for various types of accidents are shown in Table III-1. The natural retention of radionuclide vapors occurs because the vapors either condense on surfaces or react with these surfaces. Depending on the surface temperature and the duration of its exposure to high temperature steam, the surface material is either chromium oxide (Cr₂O₃) or iron oxide (Fe₃O_{4-y}). Both of these materials are expected to be reactive toward cesium-bearing vapours and strontium or barium vapors. Stainless steel lead screws above the core at Three Mile Island were found to have captured cesium by reaction with silica impurities in the steel. Metallic nickel inclusions in the oxide films on surfaces within the reactor coolant system are reactive toward tellurium whether it is in the metallic state or present as TeO or SnTe.

Aerosols, too, deposit on surfaces in the reactor coolant system by:

- gravitational sedimentation;
- impaction and interception;
- turbulent inertial deposition;
- thermophoresis; and
- under special circumstances, diffusiophoresis.

The phoretic processes, thermophoresis and diffusiophoresis, are the deposition processes likely to be susceptible to manipulation by accident management measures. They are of particular interest because unlike many aerosol processes, the phoretic processes are relatively insensitive to aerosol particle size, which will not be known well for the design of accident management strategies. Thermophoresis is the tendency for aerosol particles to move from the hot gas toward a cool surface. The rate of particle deposition is proportional to the gradient in the temperature from the gas to the surface. Accident management efforts that increase this gradient will increase thermophoretic deposition of radionuclide particles in the reactor coolant system.

Cooling to cause this increase in particle deposition can be done on the outside of the coolant system as well as by coolant additions to the reactor core. Positions that are particularly susceptible to cooling are, of course, the external surfaces of the steam generator tubes in pressurised water reactors. For this reason, accident management measures to enhance the cooling of steam generators are under consideration. Thermophoretic deposition can, however, be enhanced by cooling any of the surfaces exposed to aerosol particles in the reactor coolant system. Sustained cooling has the additional advantage of mitigating the late in-vessel source term discussed in Chapter II.

Cooling that is sufficient to condense steam can enhance aerosol particle deposition by diffusiophoresis. Diffusiophoresis is the movement of aerosol particles in a gas concentration gradient. It is a process that is usually unimportant in the reactor coolant system under accident conditions because temperatures are too high to allow much vapour to condense to create significant concentration gradients adjacent to surfaces. Vigorous cooling of surfaces as a result of accident management activities could increase the importance of diffusiophoresis. Diffusiophoresis is a particularly effective aerosol deposition mechanism because the rate of deposition is nearly independent of particle size.

Table III-1. Retention of Radionuclides in the reactor coolant system by natural deposition processes in selected accidents [V-2]

FRACTION OF INITIAL CORE INVENTORY RETAINED

ACCIDENT	Cesium	Iodine	Tellurium
PWR SBO	0.93	0.85	0.30
PWR SLOCA	0.76	0.74	0.26
BWR SBO	0.50	0.18	0.22
BWR T	0.24	0.08	0.45
PWR Large Break LOCA	0.04	0.03	0.26

LOCA = loss of coolant accident; SBO = station blackout; SLOCA = small break loss of coolant accident; T = transient initiator.

III.3 Cooling of the vessel

Currently, there is great interest in the possibility that accidents can be arrested by cooling the external surfaces of a reactor vessel. Any cooling of the core debris that can be produced this way will, of course, slow the rate of radionuclide release from core debris. The simultaneous additions of water to debris beds that accumulate within the reactor vessel will also trap radionuclides that are released. The physical phenomena involved are analogous to those involved with water overlying core debris interacting with concrete discussed in Chapter IV.

III.4 Annotated bibliography on in-vessel retention processes

V-1. A.L. Wright, *et al.*, **Primary System Fission Product Release and Transport - A State-of-the-Art Report to the Committee on the Safety of Nuclear Installations**, NUREG/CR-6193, NEA/CSNI/R(94)2, ORNL/TM-12681, Oak Ridge National Laboratory, Oak Ridge, TN, June 1994.

This report provides a comprehensive survey of experimental data and models available to predict the retention of radionuclides within the reactor coolant system during severe accidents.

V-2. R.S. Denning *et al.*, **Radionuclide Release Calculations for Selected Severe Accident Scenarios**, NUREG/CR-4624, BMI-2139, Volumes 1-5, Battelle's Columbus Division, Columbus, Ohio, July, 1986.

These reports provide a selection of accident calculations using an older computer code. Results can be of use in the definition of source term magnitudes and amounts of retention in the reactor coolant systems for the purposes of designing accident management strategies.

- V-3a. Workshop on Aerosol Behaviour and Thermal Hydraulics in the Containment, Committee on the Safety of Nuclear Installations, OECD Nuclear Energy Agency, Paris, France, 1990, CSNI Report 176.
- V-3b. Water-cooled Reactor Aerosol Code Evaluation and Uncertainty Assessment, E. Della Loggia and J. Royen, editors, EUR 11351 en, Commission of the European Communities, Directorate-General Science Research and Development and Organisation for Economic Co-operation and Development, Nuclear Energy Agency, 1988.

The proceedings of these conferences provide insight on computer codes used to predict the behaviour of aerosols in reactor containments under accident conditions. More technical details can be found in M.M.R. Williams and S.K. Loyalka, **Aerosol Science Theory and Practice** - With Special Applications to the Nuclear Industry, Pergamon Press, 1991.

IV. MITIGATION OF EX-VESSEL RELEASE

IV.1 Introduction

Reactor accidents that progress to the point that core debris penetrate the reactor vessel and cascades into the reactor containment are, of course, quite severe. At this stage in a severe accident, only the containment provides a barrier to the release of radioactive materials from the plant into the environment. Indeed, if decay heat removal from the containment could not be realized in a sufficient way, the failure of this last barrier becomes inevitable. Severe accident management efforts must, then, focus on:

- limiting containment leakage (see chapter VII);
- maintaining containment integrity; and
- attenuating the inventory of radionuclides available for release should the containment fail.

Even if the strategy to maintain containment integrity fails, this may lead to delayed containment failure. Delay in containment failure for even a few hours after core debris has penetrated the reactor vessel can have a profound effect on risk. Delay allows time for emergency preparedness measures to be put in place. It also allows time for natural and engineered processes to reduce the concentrations of airborne radionuclides within the reactor containment.

Unfortunately, it is also true that the containment could come under harsh assault at the time of vessel failure at high system pressure. This assault is then usually in the form of pressurisation and potential hydrogen combustion though in some cases it can include attack by expelled debris itself or even just elevated temperatures of the containment atmosphere. Accident management activities that can mitigate such potential threats to containment integrity are, of course, seriously considered, e.g. by primary system depressurisation in case of a high-pressure core melt sequence.

The major radionuclide release processes associated with reactor vessel penetration by core debris include:

- vaporisation and mechanical aerosol formation in case of a pressure-driven expulsion of core debris from the vessel:
- aerosol formation associated with the interactions of high temperature core debris with water outside the reactor vessel in case water is available; and
- radionuclide release and aerosol formation during core debris/concrete interactions.

These various processes that lead to radionuclide release into the containment atmosphere are discussed individually in the subsections that follow. It should be remembered that while ex-vessel releases from core debris are taking place, there can be continuing releases from residual core debris in the reactor vessel and revaporisation releases of volatile radionuclides deposited in the reactor coolant system.

IV.2 Pressure-driven debris expulsion

In case of a high pressure core melt sequence pressure-driven core debris expulsion and core debris/coolant interactions in the cavity or containment sump will produce an intense, short duration release of radionuclides to the containment atmosphere. Releases are thought to occur as a result of

vaporisation from the high temperature core debris and as a result of mechanical processes including melt entrainment in the gas and melt fragmentation by effervescing gas. Typically, aerosol produced during melt expulsion has a bimodal or even a trimodal size distribution [Ex-1].

Releases as a result of pressure driven expulsion of core debris are usually found to be risk significant only if they coincide with containment failure. On the other hand, pressure-driven melt expulsion can produce enormous quantities of aerosol (~ 400 kg of aerosol has been estimated to accompany expulsion of about 50% of a typical reactor core at operating pressure). These masses of aerosol can heavily load filter systems and affect their efficiencies at removing and retaining radionuclides in case of an early venting of the containment.

Mitigation and even elimination of the release of radionuclides by pressure-driven expulsion of melt from the reactor vessel can be achieved by reducing the pressure difference between the reactor vessel and the reactor containment. Depressurisation of the reactor vessel by either automatic means⁵ or by operator actions as an AM-measure is thought to be desirable during earlier stages of an accident so that additional water sources become available to prevent any core damage. It is usually found difficult to depressurise the reactor coolant system completely relative to the containment and to keep the system pressure at a low level during an accident progression. But, any pressure reduction helps alleviate aerosol generation by pressure-driven melt expulsion and, at pressure difference of only about 10 bar, this source is usually found to be insignificant in comparison to other sources of radioactive aerosol. Of course, as discussed in Chapter III, depressurisation of the reactor coolant system can have other consequences such as enhanced fission product release from the fuel and transport through the reactor coolant system and, perhaps, accelerated core degradation.

IV.3 Core debris interactions with coolant

Extensive release of ruthenium in case of violent fuel coolant interactions remains controversial [Ex. 2a - 2b] and though possible explanations have been provided to justify why such releases would be minimal, no experimental validation substaining the magnitude of this phenomenon currently exists. Anyway releases caused by melt interaction with water would only produce temporary increases in the containment atmosphere concentration of radioactive aerosol. So long as containment integrity is preserved, such releases are not risk significant. Again, the mass releases can cause difficulties for some filter systems because of water droplets created by the core debris interactions with coolant.

IV.4 Core debris interactions with concrete

Radionuclide release during core debris interactions and the mitigation of this release have received both analytic and experimental attention [Ex-4]. Interest in this source arises both because it can be a very important source of radionuclides in the containment atmosphere and because it can persist for many days. Tellurium release, especially, is expected to be quite protracted if mitigation steps are not taken. Release during core debris interactions with concrete occurs because the vapour pressures of most materials are considerable at the temperatures typically expected and the core debris is sparged by gases evolved from the decomposing concrete. The release of aerosols to the containment during core debris

^{5.} Most boiling water reactors have automatic depressurisation systems. Severe accidents predicted to occur at boiling water reactors often involve failure of these automatic systems to function. Improvements of the systems are being investigated.

interactions with concrete can be prolonged - persisting for days albeit at low levels. Indeed, release can continue even after the core debris has cooled to the point it solidifies. The release does contain some radioactive materials, but after a brief initial interactions with the concrete, more than 98% of the aerosol production is usually found to consist of nonradioactive materials such as concrete decomposition products and materials produced by the oxidation of metals.

The most direct process for the attenuation of radionuclide release during core debris interactions with concrete is to maintain a pool of water over the core debris. Heat transfer issues and the ability of the overlying water to quench the debris have been much debated. Most experimental evidence suggests that the attack on concrete will not be arrested by the overlaying water pool [EX-5, 6]. The same experiments demonstrate that the radionuclide release will still be sharply attenuated even though the attack on concrete persists. Attenuation of the release is thought to come about by two processes. The most obvious attenuation process is much like (though not identical) to decontamination of aerosol-laden gases that sparge through steam suppression pools. Aerosols are trapped by a combination of:

- diffusion to the walls of the bubbles;
- inertial impaction on the walls of the bubbles; and
- sedimentation within the bubbles.

Diffusion affects rather small aerosol particles. Inertial impaction and sedimentation affect larger particles. There is, then, a size of aerosol particle (nominally about $0.15~\mu m$ in diameter) that is minimally affected by the passage through the overlying water pool. Thus, the overlying water pool both reduces the magnitude of the radionuclide release during core debris interactions with concrete and changes the size distribution of particles that do reach the containment atmosphere. Particles that emerge from the overlying water pool, typically, have narrow size distribution centred around the minimally affected particle size. Particles of such sizes are frequently found to resist removal from the containment atmosphere by other processes such as spray removal.

The second effect of an overlying water pool (or due to cool-down) is to form a solidified crust at the surface of core debris interacting with concrete. This crust will certainly eliminate aerosol generation by melt entrainment in gases sparging through the core debris. It may also act to trap by interception and by impaction particles produced by vaporisation and condensation of volatile debris constituents. Quantitative assessments of the effects of the crustal material on radionuclide release during core debris interactions with concrete have not been reported.

Testing has made it apparent that core debris interacting with concrete may be quenched with water to only a short depth. The hypothesised progressive fragmentation of the core debris when it is exposed to water has seldom been observed in tests. A favoured strategy among designers now is to assure that core debris expelled from the reactor coolant system is spread thinly over a broad area so that it can be completely quenched when flooded by water. The source term consequences of spreading core debris have not been evaluated either analytically or experimentally. Certainly, core debris spreading over concrete will be vigorously sparged by the gaseous products of concrete decomposition. Sparging the molten core debris can be expected to facilitate radionuclide release from the debris. Sparging can also be expected to produce very significant quantities of nonradioactive aerosol material.

IV.5 Core retention devices

In the past and even more recently attention has been given to the use of refractory core retention devices to eliminate the release of noncondensable gases and radionuclides during core debris interactions with concrete and the attack on the reactor foundation by core debris. Typically, core retention systems involve refractory materials such as MgO and even depleted uranium oxides in place of concrete below the reactor vessel. Core debris held within these refractory materials will not be vigorously sparged by gases and, consequently, volatile constituents of the core debris including radionuclides will not be stripped from the debris. On the other hand, refractory core retention systems do lead to protracted periods in which the core debris is at temperatures high-enough that vapor pressures of radionuclides in the core debris are considerable. An alternative design for a core retention device is to use a relatively easily melted material to dilute and cool the core debris while at the same time preventing vigorous gas sparging of the core debris [Ex-7, 8].

Core retention devices are seldom found to be a permanent solution to the problem of core debris interactions. Without some mechanism for cooling the core debris, debris temperature rise to the point that the retention material is ablated at some slow rate. Eventually the retention material is penetrated and attack on structural concrete can take place. Attempts to incorporate cooling systems into the core retention system encounter significant engineering and reliability difficulties. Core retention systems, though extensively studied, have not become popular means for mitigating the consequences of severe accidents. Certainly, installation of core retention systems in existing nuclear power plants would be found usually to be an expensive, engineering challenge.

IV.6 Annotated bibliography on mitigation of ex-vessel releases

Ex-1. J.E. Brockmann, "Ex-Vessel Releases: Aerosol Source Terms in Reactor Accidents", **Progress in Nuclear Energy 19**(1987)7.

Though this paper is becoming somewhat dated, it still provides a good account about what is known concerning radionuclide release and aerosol formation during the ex-vessel stages of reactor accidents including steam explosions, pressure-driven melt expulsion and core debris concrete interactions. A good understanding of the thermal and mechanical aspects of the pressure-driven melt expulsion process and direct containment heating of dispersed core debris can be obtained from Fauske & Associates, Inc. and Sandia National Laboratories, **State-of-the-Art Report (SOAR) on HPME and DCH**, NEA/CSNI/R(96)25, OECD/GD(96)194, Committee on the Safety of Nuclear Installations, OECD Nuclear Energy Agency, Issy-les-Moulineaux, France, December 1996.

- Ex-2a. R.L. Ritzman *et al.*, "Release of Radioactivity in Reactor Accidents", Appendix VII, "**Reactor Safety Study**, NUREG 75/014, WASH 1400, U.S. Nuclear Regulatory Commission, Washington, D.C., October 1975.
- Ex-2b. D.A. Powers, "A re-examination of the Steam Explosion Source Term during Severe Accidents", p.391, **Source Term Evaluation for Accident Conditions**, International Atomic Energy Agency, Vienna Austria, 1986.

These reports discuss the radionuclide release associated with steam explosions. The original analysis in 1975 hypothesised a substantial release of ruthenium. Subsequent analysis has raised questions about a very large release of this type. There have been no experimental confirmations or refutations of either analysis.

- Ex-3a. **Proceedings of the OECD/CSNI Specialist Meeting on Fuel-Coolant Interactions**, May 19-21, 1997, NEA/CSNI/R(97), JAERI-Conf97-011, Japan Atomic Energy Research Institute, Tokai-mura, Japan, 1998.
- Ex-3b. S. Basu and T. Ginsberg, A Reassessment of the Potential for an Alpha-Mode Containment Failure and a Review of the Current Understanding of Broader Fuel-Coolant Interaction Issues, NUREG-1524, U.S. Nuclear Regulatory Commission, Washington, DC, August 1996.

These documents provide a good assessment of the current state of understanding of energetic interactions of molten core debris with water. Most of the discussion relates to energetics and the possibility of damage to the reactor. Little work has been done on the source term implications of such interactions.

Ex-4a. **Proceedings of the OECD(NEA)CSNI Specialists' Meeting on Molten Core Debris-Concrete Interactions**, KfK-5108, NEA/CSNI/R(92)10, Kernforschungszentrum Karlsruhe, Karlsruhe, Germany, 1992.

Ex-4b. Proceedings of the Committee on the Safety of Nuclear Installations (CSNI) Specialists' Meeting on Core Debris-Concrete Interactions, EPRI NP-5054-SR, Electric Power Research Institute, Palo Alto, CA, February 1987.

The proceedings of these meetings provide a good indication of the range of experimental and analytical activities devoted to the study of core debris interactions with concrete. Most of the work has dealt with concrete ablation and combustible gas generation. Relatively less work has gone into the study of radionuclide release during core debris interactions with concrete.

Ex-5. D.A. Powers and J.L. Sprung, A Simplified Model of Aerosol Scrubbing by a Water Pool Overlying Core Debris Interacting With Concrete, NUREG/CR-5901, SAND92-1422, Sandia National Laboratories, Albuquerque, NM, November 1993.

This report presents a discussion of the physical phenomena that lead to attenuation of aerosol production during core debris interactions with concrete by an overlying water pool.

- Ex-6a. R.E. Blose *et al.*, **SWISS 1 and 2: Sustained Interactions of Molten Stainless Steel and Concrete in the Presence of Water**, NUREG/CR-4727, SAND85-1546, Sandia National Laboratories, Albuquerque, NM. July 1987.
- Ex-6b. R.E. Blose *et al.*, **Core-Concrete Interactions with Overlying Water Pools**, NUREG/CR-5907, SAND92-1563, Sandia National Laboratories, Albuquerque, NM, November 1993.
- Ex-6c. B.W. Spencer, M. Fischer, M.T. Farmer, and D.R. Armstrong, **MACE Scoping Test Data Report**, ANL Report MACE-TR-D03, Argonne National Laboratory, Argonne, IL, June 1991.

Experimental demonstrations of the power of an overlying water pool to attenuate aerosol production during melt-concrete interactions even though the core debris cannot be quenched.

Ex-7. D.A. Powers, "A Survey of Melt Interactions with Core Retention Materials", **Proceedings of the International Meeting on Fast Reactor Safety Technology**, 1979.

Much of the work done on ex-vessel core retention devices has been done for fast breeder reactors. This paper describes some experimental studies of melt interactions with various materials for passive core retention devices.

Ex-8. G. Cognet *et al.*, "Core Spreading and Coolability", **Proceedings of FISA-97 - EU Research on Severe Accidents**, EUR 18258, pp.157-176, European Commission, Luxembourg, 1997.

This paper describes the new COMET system for core retention with passive cooling.

V. DECONTAMINATION OF THE CONTAINMENT ATMOSPHERE

V.1 Introduction

Over the last two decades there has been a world-wide effort to understand the behaviour of radionuclides in reactor containments under the conditions of accidents that go beyond the design bases. This effort has been spurred by major accidents at Three Mile Island and Chernobyl. This wide-ranging effort has shown that most of the important radionuclides will accumulate in the containment as aerosols. Aerosols are, of course, inherently unstable. Many naturally-occurring processes act to remove aerosols and thus decontaminate the containment atmosphere. These natural processes can be augmented by engineered systems and accident management measures to cleanse the containment atmosphere more rapidly and to reduce the inventory of radioactive material that can escape the nuclear power plant as an airborne release. Modern, mechanistic analyses show, however, that radionuclide release to the containment is protracted. Radioactive aerosols removed by various processes are replaced in the containment atmosphere by other radioactive aerosols produced by the evolving sources discussed in Chapter III. Significant concentrations of radioactive aerosol can exist in a reactor containment atmosphere for substantial periods during a severe reactor accident.

This current understanding of the behaviour of radionuclides under reactor accident conditions is, of course, quite different from the hypothesis of instantaneous appearance of radionuclides in the containment largely in gaseous form that has been used in some countries to select sites for power plants and to design safety systems for nuclear power plants. The international nuclear safety community has attempted to construct well-validated models based on the mechanistic understanding to predict the behavior of aerosols and to replace the more antiquated hypotheses. There have been several international efforts to validate the models. The LACE [A-1], Marviken-V [A-2], DEMONA [A-3] and STORM [A-4] experimental programs are important examples. The ongoing PHEBUS-FP [A-5] program is a continuation of this validation effort. Numerous computer models have been developed to describe aerosol behaviour in the containment [A-6]. Many of these have reached sufficient maturity that they are now incorporated into integrated containment response models that can be used for reactor safety analyses.

In the subsections that follow, the physical phenomena involving aerosols in reactor containments are described. The attention of the first subsection is on natural processes that decontaminate the containment atmosphere. The subsequent subsection deals with engineered systems that either attenuate the release to the containment atmosphere or decontaminate that atmosphere.

V.2 Natural aerosol processes in the containment

Aerosols in reactor containments are unstable. They tend to grow in size and to deposit on surfaces. Given sufficient time, the atmosphere of a containment building will naturally cleanse itself to an important extent. Aerosols deposit from the atmosphere by several mechanisms of which the most important are:

- gravitational settling;
- turbulent deposition on surfaces;
- thermophoresis which is the movement of particles down the temperature gradient between the hot atmosphere and cooler surfaces; and
- diffusiophoresis which is the movement of particles down the concentration gradient created by the condensation of steam on a surface.

Deposition of particles in a reactor containment will usually result in the particles being trapped in water. This trapping is usually thought to result in permanently removing the particles and its associated radioactivity from the inventory of materials that can leak from the containment or be released suddenly from the plant should the containment rupture. Experiments and analyses have shown that even vigorous "steam flashing" from hot water pools in the event of rapid containment depressurisation is insufficient to resuspend substantial fractions of the radioactive particles trapped in water. (Iodine is an exception to this generalisation and the exceptional behaviour of iodine is discussed in Chapter VI.)

2.1 Particle growth and gravitational sedimentation

In large containment volumes found at many existing nuclear power plants, gravitation settling of aerosols is the predominant mode of natural decontamination of the atmosphere. Gravitational settling rates increase with the increases in the size of the aerosol particles. Particles suspended in the containment atmosphere grow to sizes that will sediment at significant rates by two mechanisms:

- coagulation with other suspended particles; and
- condensation of steam on the particles.

Coagulation of particles is, by far, the more important of these processes. Coagulation rates depend strongly on the concentration of particles in the containment atmosphere. Both nonradioactive and radioactive particles contribute to the coagulation of all particles. It is for this reason that releases of nonradioactive materials during core degradation and accident progression are of nearly as much interest as are the releases of radionuclides. The nonradioactive particles can accelerate the growth and gravitational sedimentation of radioactive materials. Nonradioactive aerosol materials must also be included in design loads for engineered systems that augment natural aerosol removal from the containment atmosphere.

Coagulation of aerosol particles occurs when particles cross streamlines of flow and come into contact. Such contacts between particles can be caused by:

- rapid settling of larger particles sweeps out slower, smaller particles;
- Brownian movement of particles across streamlines; and
- turbulent diffusion and turbulent inertia move particles across streamlines.

Gravitational collisions between particles can be quite an important mechanism of coagulation in reactor containments because of the nature of the aerosol particle size distribution that develops in the containment atmosphere. Such size distributions are often predicted to be bimodal when there is an operating source of aerosol to the containment. The small size mode of the bimodal distribution is produced by particles freshly injected into the atmosphere from the operating source. The larger size mode is made up of particles that have "aged" in the atmosphere. Once sources of aerosol to the containment become small, the size distribution quickly becomes unimodal.

Turbulent coagulation processes only modestly accentuate the rates of particle growth. Turbulent coagulation is usually characterised in terms of the turbulent energy dissipation rate in the atmosphere. Relatively crude models now available for predicting this turbulent energy dissipation rate are probably adequate for estimating the incremental increases in particle growth rates caused by turbulence. Accident management efforts that accentuate particle growth by increasing the level of turbulence might necessitate the development of superior containment hydraulics models.

Aerosols in a reactor containment will be radioactive and subjected to an intense ionising radiation field produced by the radioactive noble gases. In the past, it had been assumed that any electrostatic charging of aerosol particles as a result of their radioactivity would be neutralised by fluxes of gaseous ions formed in the radiation field. This neutralisation will occur only if the mobilities of positively and negatively charged ions in the containment atmosphere are approximately equal. If this is not the case, and it isn't the case in the earth's atmosphere, electrostatic forces between particles will affect coagulation. Models suitable for the evaluation of the effects of electrostatic forces on coagulation and particle deposition in reactor containments have not been reported. An experimental study of the effects of electrostatic forces is being undertaken. Models of the effects may be of use in the design of electrostatic deposition strategies for the control of accident source terms.

Other phenomena can accentuate the agglomeration of aerosol particles and, thus, accentuate the rates of particle removal from a containment atmosphere. Agglomeration of particles by sonically driven processes has been considered as an accident management measure. Agglomeration can occur in a standing sound wave in a reactor containment because there is a phase lag between the responses of particles of different size to the gas vibrations.

Particle growth by the condensation of steam has been an area of some controversy. Were the aerosol particles in containments totally inert, steam condensation would not be an issue of great significance. Steam condensation causing particle growth would occur only when the containment atmosphere was supersaturated - a condition that would not long endure. But, aerosol particles need not be inert. They may, in fact, contain constituents that are quite hygroscopic. For instance CsOH which is often cited as a chemical form of cesium to be expected in a containment atmosphere is one of the most hygroscopic materials known. This hygroscopicity will lead to steam condensation on the particles even at low relative humidities. It has proven difficult to include the effects of hygroscopicity in models of aerosol behaviour because this effect depends so strongly on the particular chemical species present in aerosols. For example, cesium may be present in the containment atmosphere as the very hygroscopic material CsOH or as the much less hygroscopic material Cs₂ZrO₃. Iodine may be present as the very hygroscopic material CsI or as the virtually nonhygroscopic material AgI. Furthermore, the aerosol particles will not be pure materials with well-established properties. Hygroscopic chemical forms will quickly agglomerate with nonhygroscopic materials and the properties of the resulting mixture are difficult to predict with any confidence. Detailed information about the aerosol particles produced in reactor accidents that would be needed to make an acceptably accurate prediction hygroscopicity is not likely to be available soon.

Calculation of gravitation sedimentation rates is usually based on the assumption that the containment atmosphere (or, more recently, the individual compartment atmosphere) is well mixed so that the particle concentration is spatially uniform. This need not be the case. Instances where stratification of the atmosphere is possible at least in particular compartments of containments have been identified. More sophisticated models would be needed to predict accurately the gravitational sedimentation of particles from these poorly mixed atmospheres. Accident management measures may, in fact, be used to prevent the development of stratified regions from which radioactive aerosol are removed slowly.

Sedimentation velocities of aerosol particles depend on particle shape. The models of aerosol behaviour that are now available are derived for perfect spheres that have no porosity. The deviations of real particles from this ideal are handled by correction factors called shape factors. In the case of gravitational settling, the dynamic shape factor is used to account for deviations from sphericity and for porosity. These shape factors are not known well and frequently are estimated by back calculation from experimental data for simulant aerosols. This, of course, is not a reliable procedure. There have been some attempts to predict shape factors based on the fractal nature of particles that have grown by coagulation [A-7b].

2.2 Turbulent deposition

Turbulent deposition of particles on surfaces becomes an important process in reactor containments only when particle concentrations have become small. Accurate modeling of turbulent deposition rates has been a topic of debate for some time within the aerosol science community. Models currently available are usually assumed to be adequate in light of the relatively crude understanding of turbulent hydraulics within reactor containments that is now available. These models are based on the study of turbulent flows through pipes. The models are based on the hypothesis that turbulent impulses to aerosol particles can thrust particles across laminar boundary layers adjacent to structural surfaces if the layers are thinner than the 'stopping distance' of the particles.

2.3 Phoretic deposition of particles

Thermophoretic and diffusiophoretic deposition processes arise because of the noncontinuum nature of the containment atmosphere at the length scales of aerosol particles. In simplistic terms, gradients in temperature or concentration make impulses imparted by gas molecules on one side of an aerosol particle different from impulses imparted to the other side. There is, then, a net force on the particle that causes the particle to accelerate until the phoretic force is balanced by the drag force on the particle. This leads to a steady-state velocity of the particle down the gradients in temperature and concentration. The deposition by these phoretic processes is very interesting for accident management because particle deposition velocities are not especially sensitive to particle size. The combination of a thermal gradient and a concentration gradient created by the condensation of steam can lead to efficient particle deposition. Large gradients in concentration or temperature are not predicted usually to long endure within containments of existing nuclear power plants under accident conditions. Phoretic processes do not provide, then, continuing methods of efficient aerosol removal from the atmosphere. Prolonged periods of high phoretic deposition need to be engineered into the containment. Cooled containment surfaces being considered for heat removal in advanced reactor designs can also provide efficient aerosol removal.

V.3 Engineered safety systems

Natural processes do not decontaminate containment atmospheres rapidly. Decontamination rates are often characterised in terms of the decontamination coefficient, λ , which indicates the reciprocal of the time required to reduce the aerosol concentration by a factor of $1/e \approx 2.7$. Typical values for λ due to natural processes in a large dry containment are less than 0.5 hr^{-1} . Such slow decontamination may not provide sufficient protection when large threats to containment integrity exist. Some engineered systems are available in containment that can accelerate the rates of aerosol particle removal. In general, these systems were not included in the containment design for aerosol particle removal. They were designed to

reduce steam pressurisation of containment following a large break in the reactor coolant system. They are, then, better suited to handle a large event of short duration than the prolonged release of radioactivity in a severe reactor accident. The processes involved in particle removal by these engineered systems are of interest since similar process and systems could be used in accident management strategies. In fact, one element of an accident management strategy could be to preserve the capabilities of these engineered systems to decontaminate the atmosphere over the prolonged period of a reactor accident.

3.1 Containment sprays

The quintessential engineered system for the removal of aerosol particles is the spray found in the containments of many pressurised water reactors and in the drywells of many boiling water reactors. Sprays remove particles by:

- sweepout of particles unable to avoid the falling droplet;
- interception of particles as they follow steamlines of flow around falling droplets; and
- diffusion of aerosol particles to the droplet surface.

Sweepout and interception most efficiently remove larger particles (particle diameter $> 1~\mu m$). Diffusion is most efficient for very small particles (particle diameter $< 0.1 \mu m$). Consequently, very fine and very large aerosol particles are efficiently removed from the containment atmosphere by sprays. There is, however, an intermediate size of particle that is minimally affected by sprays. The decontamination of the atmosphere of these intermediate sized particles is increased by decreasing the size of the spray droplets, which are typically between 250 and 2000 μm in diameter. Because of the particle size dependence of spray effectiveness, the spray not only changes the concentration of particles in the atmosphere, it also changes the size distribution of these aerosols. The residual aerosol has a narrow distribution of sizes centered around the size minimally affected by the spray.

Sprays can be tremendously efficient in particle removal. Values of λ in excess of 10 hr⁻¹ can be produced by typical sprays in reactor containments. The efficiency of the spray at particle removal does decrease with time as the remaining aerosol particles are concentrated in the size range that is minimally affected by the spray. Spray efficiency is also reduced when there are large unsprayed volumes within the containment with poor hydraulic communication with the sprayed volumes. This limitation on spray effectiveness is usually the reason that sprays designed for fire protection are often found to be inefficient accident management tools. Sprays can also require the availability of AC power. One common objective of accident management strategies is to find ways to actuate containment sprays in accidents like station blackout accidents where the usual sources of water and power for the sprays are not useable. Sprays that can be driven passively by gravity or by pressurised gas have become of interest for accident management.

3.2 Steam suppression pools

Gases laden with radioactive particles in accidents in boiling water reactors can be directed through the steam suppression pool [A-8]. Effluents from the reactor coolant system can pass through so-called X-quenchers or T-quenchers into the pool. These quenchers are submerged 30 cm diameter pipes with many 1-2 cm diameter holes through which gas can sparge the suppression pool. Effluents from the drywell are directed into the pool via very large diameter (~60 cm) pipes. Flows into the pool are broken into bubbles. Aerosols are removed from the bubbles by:

• condensation of supersaturated steam;

- sedimentation within the bubble;
- diffusion of particles to the bubble surface;
- inertial impaction of particles with bubble surfaces; and
- sweepout of particles by bubble oscillations.

Particle fluxes to the bubble walls can be resisted by the evaporation of water vapour into the bubble as it rises through the pool and loses hydrostatic head. This resistance by evaporation becomes more significant as the pool temperature increases toward saturation. Circulation of gases within a rising bubble that leads to inertial impaction of particles can be damped by accumulation of surface-active impurities on the bubble surface.

Aerosol removal from bubbles is very dependent on bubble size. Removal is more efficient from smaller bubbles. Fortunately, bubbles rising in a suppression pool disintegrate to a common size of about 0.5 cm regardless of how they are injected into the pool [A-9].

Aerosol removal processes that occur when a bubble rises through a suppression pool vary in efficiency with particle size. As with sprays, very fine and very large particles are efficiently removed. There is a particle size that is minimally affected by the decontamination processes. Aerosols that emerge from a suppression pool have sizes narrowly distributed around the minimally affected particle size (also called the maximum penetration size). These residual aerosols also resist removal by many other decontamination processes so they can be quite persistent in the atmosphere.

Aerosols are removed during the process of formation of bubbles in the suppression pool as well as during bubble rise through the pool. Particles in the gas jet that forms the bubble can impact the developing bubble wall or diffuse to the bubble surface. Recent analyses suggest that particle removal during bubble formation may be comparable with particle removal during bubble rise [A-9].

Aerosol removal by suppression pools has received quite a lot of experimental and analytical attention. Computer models of the removal process include the SPARC model [A-10a], the BUSCA model [A-10b], and the proprietary model SUPRA [A-10c]. Excellent experimental studies have been conducted [A-11] and additional studies are underway in Europe and Japan [A-9a, b].

Similar aerosol removal can be expected in VVER-440 reactors equipped with bubbler condensers [A-12].

3.3 Fan coolers

Some pressurised water reactor containments are equipped with fan coolers. These coolers would appear to offer the possibility of particle deposition by thermophoresis or even by diffusiophoresis. Analyses of the potential decontamination that could be achieved with such fan coolers have not been reported. They may not provide a long-term aerosol removal capability. Accumulation of insulating deposits of aerosol on their surfaces may limit effectiveness. Fan coolers do require power to operate and the necessary power may not be available under accident conditions.

3.4 Effects of combinations of engineered safety systems

Engineered safety systems can produce very significant decontaminations of containment atmospheres. It is tempting, then, to hope that the combination of two engineered systems would produce extraordinarily rapid and complete decontamination. Seldom is this true. Nearly all engineered systems produce decontamination that depends on the aerosol particle size. Usually the very large and very small particles are removed with great efficiency and particles of intermediate size (0.1 to 0.5 µm diameter) are not removed very well at all. A mass of aerosol whose size distribution has been altered during decontamination by one system to consist primarily of particles that are only poorly removed by the system will also defy extensive decontamination by other systems unless special design precautions are taken. Thus, the residual aerosol that has passed through a steam suppression pool will not be further decontaminated substantially by a spray. Furthermore, because the residual aerosol particles are of similar size, agglomeration will increase the sizes of the particles quite slowly. The advantages of multiple systems for decontaminating containment atmospheres are nearly always found to be the reliability advantages of redundancy and not the advantages of significantly greater decontamination.

- V.4 Annotated bibliography of aerosol removal from the containment atmosphere
- A-1. F.J. Rahn, Summary of the LWR Aerosol Containment Experiments (LACE) Program Interim Report, LACE TR-012, Electric Power Research Institute, Palo Alto, CA, January 15, 1987.
- A-2. J. Collen, H. Unneberg, and D. Mecham, "Overview of Marviken Experimental Procedures", Proceedings of the ANS Meeting on Fission Product Behavior and Source Term Research, NP-4133-SR, paper 20-1, Electric Power Research Institute, Palo Alto, CA, 1985.
- A-3. M. Eusebi, *et al.*, "Preparatory Calculations for a New Experimental Program on Dry Aerosol Resuspension Mechanisms (STORM Project)," **Proceedings of the European Aerosol Conference**, Oxford, United Kingdom, 1992.
- A-4. P. von der Hardt and A. Tattegrain, "The PHEBUS Fission Product Project," **Journal of Nuclear Materials**, *188*(1992)115.
- A-5. T.K. Kanzleiter, **DEMONA Experiments Final Report**, BIeV R65.523-01, Battelle Institute E.V., Frankfurt am Main, Germany, September 1987.

These papers describe some of the experimental research on which our understanding of aerosol behavior in reactor containments is based.

- A-6a. Workshop on Aerosol Behavior and Thermal Hydraulics in the Containment, Committee on the Safety of Nuclear Installations, OECD Nuclear Energy Agency, Paris, France, 1990, CSNI Report 176.
- A-6b. Water-cooled Reactor Aerosol Code Evaluation and Uncertainty Assessment, E. Della Loggia and J. Royen, editors, EUR 11351 en, Commission of the European Communities, Directorate-General Science Research and Development and Organization for Economic Cooperation and Development, Nuclear Energy Agency, 1988.

The proceedings of these conferences provide insight on computer codes used to predict the behavior of aerosols in reactor containments under accident conditions. More technical details can be found in M.M.R. Williams and S.K. Loyalka, **Aerosol Science Theory and Practice** - With Special Applications to the Nuclear Industry, Pergamon Press, 1991.

- A-7a. Nourbakhsh, S.E. Perez and J.R. Lehner, **Effectiveness of Containment Sprays in Containment Management**, NUREG/CR-5982, Brookhaven National Laboratory, Upton, NY, May 1993.
- A-7b. D.A. Powers and S.B. Burson, A Simplified Model of Aerosol Removal By Containment Sprays, NUREG/CR-5966, SAND92-2689, Sandia National Laboratories, Albuquerque, NM, June 1993.

These documents provide descriptions of the physical processes associated with spray removal of aerosols from containment atmospheres and the expected efficacy of removal processes under accident conditions.

- A-8a. K. Fischer and W. Häfner, **Retention of Aerosols in Water Pools**, BF-V38.070-01, Battelle Engineering Corp., Frankfurt am Main, Germany, March 1994, and
- A-8b. D.A. Powers, **A Simplified Model of Decontamination by BWR Steam Suppression Pools**, NUREG/CR-6153, SAND93-2588, Sandia National Laboratories, Albuquerque, NM, May 1997.

These documents provide thorough discussions of the physical and chemical phenomena that affect the removal of aerosols from gases sparging through water pools.

- A-9a. M.J. Marcos, F.J. Gomez, I. Melches, M.M. Espigares, and J. Lopez, **LACE-ESPAÑA Experimental Programme on the Retention of Aerosols in Water Pools**, GE-93/03, ITN/TS-08/DP-93, 1993.
- A-9b. J. Hakii, H. Kaneko, M. Fukasawa, Y. Masahiro, and M. Matsumoto, "Experimental Study of Aerosol Removal Effect by Pool Scrubbing," **Second Workshop on LWR Severe Accident Research at JAERI**, Tokyo, Japan, 1991.
- A-9c. K. Hashimoto, K. Soda, and S. Uno, "High Pressure Pool Scrubbing Experiment for a PWR Severe Accident," **Proceedings of the ANS International Topical Meeting**, Portland, Oregon, July 1991.

Experimental programs that are intended to validate the models of aerosol removal by steam suppression pools and other water pools.

- A-10a. P.C. Owczarski and K.W. Burk, SPARC-90: A Code for Calculating Fission Product Capture in Suppression Pools, NUREG/CR-5765, PNL-7723, Pacific Northwest Laboratory, Richland, WA, October 1991.
- A-10b. S.A. Ramsdale, **BUSCA-JUN90 Reference Manual**, SRD R542, Safety and Reliability Directorate, United Kingdom, 1991.
- A-10c. A.T. Wassel, A.F. Mills, D.C. Bugby, and R.N. Oehlberg, "Analysis of Radionuclide Retention in Water Pools", **Nuclear Engineering and Design**, *90*(1985)87.

These are the major computer models for predicting the decontamination of aerosol laden gas streams sparging through water pools.

A-11. D.D. Paul, L.J. Flanigan, R.A. Cudnik, and J.C. Cunnane and R.P. Collier, **Radionuclide Scrubbing in Water Pools, Volume 1: Gas-Liquid Hydrodynamics**, NP-4154, Volume 1, Battelle Columbus Laboratory, Columbus, OH, August 1985.

This is a report on an outstanding experimental study of the behavior of bubbles injected into pools through quencher orifices.

A-12 H. Karwat, Status of the Bubbler Condenser Containment System for the Reactors of the VVER-440/213 Type, OECD Nuclear Energy Agency, Issy-les-Moulineaux, France, 1998, NEA/CSNI/R(98)13.

VI. MITIGATION OF GASEOUS IODINE

VI.1. Introduction

Discussions in this chapter are on the chemistry of iodine and the mitigation measures that can be taken to reduce the potential for iodine release from a containment following a reactor accident. Iodine is expected to be released to the containment atmosphere during a reactor accident predominantly as aerosols containing metal iodides such as CsI, AgI, InI, FeI₂, etc. and some small fraction of gaseous iodine (HI, I, and I₂(g)). During the early stages of a reactor accident, the inventory of iodine suspended in the containment atmosphere is determined by the releases of these aerosol and gaseous forms of iodine. Some conversion of iodine from one form to the other because of vapor interactions with aerosols can take place. Hydrogen combustion events in the containment atmosphere could cause iodine in aerosol form to be converted to the gaseous form and this possibility is discussed in this chapter. But, the most important process taking place during the earlier phases of the accident is the removal of iodine in both gaseous and aerosol forms from the atmosphere. Removal may be by engineered safety systems such as containment sprays or steam suppression pools or it may be by the action of natural deposition processes of aerosols and vapours. These removal processes will, typically, result in iodine being accumulated in the bodies of water within the containment. The peculiarity of iodine chemistry is that if iodine can dissolve in water, it can undergo chemical transformations to a volatile chemical form such as I₂(aq) or an organic iodide such as CH₃I(aq). The volatile chemical forms of iodine can partition back into the containment atmosphere. That is, retention of iodine in the containment water bodies need not be permanent.

The ability of iodine to partition from water into the containment atmosphere is significant in terms of the potential consequences of a reactor accident. Partitioning assures that for many days following an accident there can be some concentration of iodine vapour in the containment atmosphere. This iodine in the atmosphere is available to leak from an intact containment or to be released in the event of catastrophic containment failure. Partitioning of iodine from water into the gas phase can also be a mechanism for radioactive iodine to bypass filtering systems that are quite effective at mitigating the releases of other radionuclides from reactor containment. It is the persistence of iodine suspended in the containment atmosphere more than the chemical or physical form of iodine that raises risk-significant issues.

Interest arises, then, in the management of the potential iodine source term in a reactor accident. The interest is either to mitigate the tendency for iodine to partition into the atmosphere from water or to remove the iodine that reaches the atmosphere. The processes that can affect iodine partitioning (Section VI.2.1) and the processes that can affect the containment atmosphere concentrations of iodine (Section VI.2.2) are discussed in this chapter. Computer codes that may be used to assess the threats posed by the iodine source term and the effectiveness of mitigation measures are discussed in Section VI.3. Processes involved in the filtration of iodine from gases released from the containment are discussed in Section VI.4. Methods for managing the iodine source term are summarized in Section VI.5. Current practices within the nuclear community for managing and controlling the iodine source term are presented in Section VI.6. Finally, an annotated bibliography that provides sources for more detailed information pertinent to the management of the iodine source term is presented in Section VI.7.

VI.2 Iodine chemical and transport behaviour within containment

Iodine dissolved in water will be present predominantly as the nonvolatile iodide ion, I , under most conditions. Here, the interest is in the transformation of the iodide ion into a volatile chemical form, the partitioning of this volatile chemical form into the atmosphere, and the behaviour of the volatile chemical form in the atmosphere. It will be shown that the processes taking place in the containment involving the volatile chemical forms of iodine result in re-trapping the iodine as iodide ion in water. A sort of dynamic equilibrium is established in the containment involving:

Non-Volatile Iodine
$$\rightarrow$$
 Volatile Iodine

Volatile Iodine
$$\rightarrow$$
 Non-Volatile Iodine (2)

This dynamic equilibrium results in a steady-state concentration of gaseous iodine in the containment atmosphere. The objective of iodine management following a reactor accident is to reduce this steady-state concentration. The mechanisms for reducing the concentration in the containment atmosphere require an appreciation of the aqueous chemistry of iodine and the reactions of iodine vapours in the containment atmosphere. These topics are the foci of discussions in this subsection. From these discussions arise a number of approaches for mitigating the atmospheric concentration of iodine that are discussed further in the subsequent subsections of this chapter.

2.1 Aqueous phase reactions of iodine

Iodine in water can be present in a number of chemical forms including the familiar iodide ion, Γ , the volatile form, $I_2(aq)$, and the highly oxidised iodate ion, IO_3 , as well as a number of transient forms with oxidation states intermediate between those of the more familiar forms. Even under normal conditions, there is some tendency for interconversions among these forms, but the conversion processes can be glacially slow under normal conditions. A radiation field can greatly accelerate the formation of various chemical forms of iodine in water. Products of water radiolysis can oxidise the iodide ion to form molecular iodine following a reaction scheme that is summarily described by:

$$\Gamma + \bullet OH \rightarrow I \bullet + OH^{-}$$
 (3)

$$I_{\bullet} + I_{\bullet} \rightarrow I_{2}(aq)$$
 (4)

The production of the key reactant, the hydroxyl radical (•OH), increases with increases in the radiation dose rate to the water. The dose rate to the water depends, of course, on the total amount of fission products and other radionuclides that escape the core and enter the containment. Dose rates to sump waters will vary depending on the type of reactor, the type of accident and with time following an accident. Typical values will be in the range of 1 to 20 kGy hr⁻¹.

The molecular iodine in water can disproportionate and other products of water radiolysis can reduce it back to iodide:

$$I_2(aq) + H_2O \rightarrow HOI + I^- + H^+$$
 (5)

$$I_2(aq) + O_2^- \rightarrow I_2^- + O_2(aq)$$
 (6)

$$I_2(aq) + H_2O_2 \rightarrow 2I^- + 2H^+ + O_2(aq)$$
 (7)

These reactions that destroy aqueous, molecular iodine depend strongly on the pH of the solution. The rates of reactions (5) and (7) increase significantly with temperature. Because the of these reactions that destroy $I_2(aq)$ and the reactions of iodide with radiolysis products that form $I_2(aq)$, a steady state concentration of this volatile form will develop in the solution.

The steady-state concentration of $I_2(aq)$ in the solution will depend on a subtle balance between the rates of chemical reaction that form volatile iodine and the rates of chemical reactions that destroy volatile forms of iodine. The concentration of volatile iodine in solution will determine the concentration of molecular iodine in the containment atmosphere by way of the equilibrium:

$$I_2(aq) \leftrightarrow I_2(g)$$
 (8)

The equilibrium constant for this process is usually presented in the form of a partition coefficient, H, which is defined as the ratio of the concentration in the aqueous phase to the concentration in the gaseous phase at equilibrium:

$$H = partition coefficient = \left(\frac{concentration in the aqueous phase}{concentration in the gas phase}\right)$$
(9)

Note that the smaller the partition coefficient, the higher the concentration in the gas phase relative to the concentration in the aqueous phase. In water at room temperature, the partition coefficient of molecular iodine is about 80.

In addition to the reactions in solution mentioned above, molecular iodine dissolved in water can react with submerged surfaces. Irreversible chemical adsorption of iodine species on submerged surfaces could reduce the overall iodine inventory in the aqueous phase, and thereby reduce the release of molecular iodine to the containment atmosphere.

Surfaces exposed to solutions in the containment may be painted or bare steel. Most painted surfaces or clean steel surfaces in contact with water are not good iodine absorbers. Several studies have shown, however, that the zinc primer used to protect carbon steel in containment from corrosion can absorb iodine effectively from solutions at high pH (pH \sim 9-10). The detailed mechanism of iodine absorption on zinc primer surfaces has not been identified. It has been proposed that the metallic zinc in the coating is oxidised by dissolved oxygen in water to form various divalent, insoluble complexes, and that iodide ion, I $^-$, may be incorporated in these complexes. The solubilities of the divalent zinc complexes depend on pH and the minimum solubilities are in the range of pH = 9.3 to 9.7. At higher and lower values of pH, reactions of iodide ion with zinc primers would be less effective at mitigating the production of molecular iodine.

A metal with great affinity for iodine is silver. The importance of the reaction of metallic silver with iodine was first demonstrated in the LOFT FPT-2 test. Renewed attention to the reaction of molecular iodine with silver has arisen based on the results of the PHEBUS FPT0 and FPT1 tests in which silver-indium-cadmium control rod materials were released into the containment model of the test facility. Iodine appeared to react with this silver to form water-insoluble silver iodide (AgI). Precipitation of AgI appeared to control the behaviour of iodine in the containment model. Though details of the interactions of molecular iodine and iodide ion with metallic silver are still being investigated, it appears that silver in contact with water can be a very effective adsorber of iodine even at low values of pH. Deliberate addition of silver into containment sumps to augment any silver released to the containment by accident processes might be used to manage the iodine source term. For silver to retain iodine, the surface must remain immersed in water. Silver iodide on a silver surface exposed to air is rapidly oxidised to form silver oxide with the release of molecular iodine. Also, silver iodide may react with sulphur compounds in the atmosphere to form silver sulphide and release iodine. Finally, there is evidence that silver iodide is not stable to the beta radiation produced by radioactive iodine.

As if this chemistry of iodine in water were not complicated enough, there is also the possibility of aqueous iodine in water and a radiation field reacting to form volatile organic iodides:

$$RH(aq) + \bullet OH \rightarrow R \bullet + H_2O$$
 (10)

$$R + I_2(aq) \rightarrow RI(aq) + I_{\bullet}$$
 (11)

where the symbol R is used to indicate a general organic radical. Most familiar of the organic iodides that can formed by this process is methyl iodide, CH₃I, though by no means is this the only or even the most likely organic iodide that can be formed under accident conditions. Formation of organic iodides depends on the availability of organic reactants. A major source of water-soluble organic compounds such as ketones and alcohols in a reactor containment is likely to be the solvents used in paints for containment surfaces. Freshly painted surfaces (aged for 3 months) in containment can release as much as 0.01 moles organic per square decimeter (1x10⁻² mol dm⁻² either directly to the water if immersed or to the atmosphere. The amount of organic solvent trapped in painted surfaces will decrease with the age of the paint due to gradual loss by evaporation, but even 5 year old paint is observed to release as much as 1×10^{-4} mole organic dm⁻². Of course, water-soluble organic species will partition between the atmosphere of containment and the aqueous phase. Another source organic material may be the radiolytic breakdown of oil and grease that releases small, soluble organic compounds. Again, radiolytic breakdown of organic materials can release water-soluble organic materials directly to the aqueous phase or to the containment atmosphere. Consequently, it usually assumed adequate organic will be available in reactor containment to sustain reactions that lead to organic iodide formation.

In hot water and a radiation field, the organic iodides can decompose:

$$RI(aq) + H_2O \rightarrow I^- + H^+ + ROH(aq)$$
 (12)

$$RI(aq) + OH^{-} \rightarrow I^{-} + ROH(aq)$$
 (13)

Again, because of the simultaneous formation and destruction processes, a steady state concentration of organic iodide can develop in solution and this organic iodide can partition into the atmosphere:

$$RI(aq) \leftrightarrow RI(g)$$
 (14)

Partition coefficients for some organic iodide species are smaller than those for $I_2(aq)$. The partition coefficient for CH_3I , for example, at room temperature in water is about 5. On the other hand, other organic iodides can have partition coefficients much higher than that of molecular iodine in water. The partition coefficient of 2-iodophenol, for example, is about 725. Recent experimental studies indicate that organic iodides other than CH_3I with higher partition coefficients than CH_3I could be formed abundantly during severe reactor accidents. Accident management activities to control the release of iodine from a reactor containment following an accident must consider the formation of organic iodides as well as the formation of molecular iodine. It appears, however, that focusing on the control of molecular iodine, $I_2(aq)$, is sufficient to also control formation of organic iodides.

2.2 Key parameters affecting net volatile iodine production

The production of molecular iodine in the aqueous phase involves the relative balance between reactions that oxidise iodide ion and reactions that reduce molecular iodine. This balance may be affected the radiation dose rate, which is beyond control by accident management measures, and by the many chemicals introduced during accident progression. The balance between oxidation and reduction is also affected by temperature and by the pH of the solution. The temperature dependence of the molecular iodine concentration in solution is complicated. Reactions that oxidise

iodide ion or lead to the formation of organic iodides are fast free radical reactions. Rates of these reactions are not especially sensitive to temperature. Many of the reactions that reduce molecular iodine or hydrolyse organic iodides are thermal reactions whose rates are often found to increase significantly with increasing temperature. Thus, increasing temperature should shift the balance between formation and destruction of molecular iodine and organic iodides in solution toward destruction. On the other hand, the partition coefficients of the volatile species decrease with increasing temperature. This decrease in the partition coefficient increases the equilibrium concentration of volatile iodine in the atmosphere relative to the concentration in the aqueous phase and thus offsets in part the lower concentration of volatile iodine in the aqueous phase at elevated temperatures. It is now thought that iodine release to the ambient gas phase may pass through a maximum at temperatures of 80 to 90 °C. Attempts to control water temperature as a means to control iodine release to the containment atmosphere appear to be difficult and are likely to have limited effectiveness.

Much more profound is the sensitivity of molecular iodine formation to the solution pH. The aqueous concentration of molecular iodine will decrease by about an order of magnitude with an increase of the pH by one unit in the pH range of five to ten. Therefore, maintaining the pH of water in the containment at a high value following an accident is thought to be a practicable method for mitigating the formation of molecular iodine.

The pH of solutions can be controlled to high values by the addition of strong base or by the addition of a buffer. Design of a strategy for managing the iodine source term does require some appreciation for the factors that will affect pH over the course of an accident. The initial pH of solutions in the containment is dependent on reactor design. Different reactor designs maintain the pH of their heat transport coolant, their emergency core coolant and containment dousing water at different values, some of which are quite basic (high pH). Factors that cause changes in this initial pH over the course of a reactor accident are of a more generic nature. Here, the factors considered are: (1) nitric acid or NO_x formation by the radiolysis of moist air, (2) the formation of inorganic acids such as HCl by the radiolysis and pyrolysis of organic materials in the containment, (3) organic acid formation from the radiolysis of organic materials, and (4) calcium hydroxide leached from the containment and CaO, Na_2O , and K_2O aerosols formed during interactions of core debris with concrete.

Nitric acid can be formed by the radiolysis of moist air. This nitric acid will rapidly dissolve in water and cause a decrease in the pH. On the other hand, intermediate products in the air radiolysis process interact rapidly with surfaces and other chemical species in the atmosphere. Furthermore, nitric acid is susceptible to decomposition in a radiation field. It has been difficult, then, to assess the overall effect nitric acid formation by radiolysis will have on solution pH.

Experimental studies have shown that irradiation and heating of polyvinyl chloride insulation found on electrical cables in many reactor containment will yield both hydrochloric acid (HCl), sulfurous acid (H_2SO_3), and sulfuric acid (H_2SO_4). Some investigators believe that acids formed by the combination of radiolysis and pyrolysis of electrical cable insulation can be of dominant importance to the pH of solutions in the reactor containment. Others feel that acid formation by this process is over-emphasized because acids generated within the polymer are not able to escape to the atmosphere except when they are formed near the polymer surface.

Recent experimental studies have lead some investigators to argue that the radiolytic formation of organic acids such as acetic acid and formic acid may be the most important drivers for changing the pH of water in reactor containment during the course of severe accidents. The many sources of organic materials that can be broken down by irradiation were mentioned above. There are organic paints, cable sheathing, and lubricants within containment that are susceptible to radiolysis. The products of organic decomposition that are soluble in water are of most interest. Long chain, organic molecules and nonpolar alkyl-organic compounds that can be formed by pyrolysis and radiolysis of organic polymers are not very soluble in water and are not likely to react to form organic acids. Ketones and alcohols are more likely to react to form acids that will affect the pH of water in the containment.

Finally, mention must be made of the effects of other materials released into water in the containment over the course of an accident. Some of these materials are released to the containment during core degradation. The effects of these materials on water pH are not well understood. Hot water will leach calcium hydroxide from concrete and this can cause an increase in pH. Core debris interactions with concrete release copious quantities of aerosol that are rich in species like CaO, Na_2O and K_2O that will dissolve in water to form hydroxide ions and raise the pH. On the other hand, CaO can precipitate buffers intended to control the water pH.

Several chemicals have been considered as base additives or buffering agents for maintaining a high pH in the sump water. When choosing chemicals for use as a buffering agent (and/or a base additive) for this application, factors that need to be considered include thermal stability and stability over a range of dew points during storage, stability in the presence of radiation following an accident, and buffer capacity (or base concentration) at high pH values. For example, hydrazine, an additive considered for spray water systems to scavenge oxygen to prevent corrosion, is a base which can increase pH. But, hydrazine decomposes rapidly in the presence of radiation and thus continuous injection of this chemical would be required for accident mitigation. If a base is used without a buffer, the initial base or OH concentration must be larger (by at least an order of magnitude) than the total expected H⁺ production during an accident.

Tri-sodium phosphate and borate/boric acid have been identified as good candidates for containment sump pH control based on the above considerations. They have high buffer capacities at high pH values (i.e., high pKa values) and, being solid, are easy to handle. Phosphate has two useful buffer regions:

$$H_2PO_4^- \leftrightarrow HPO_4^{2-} + H^+ \longleftrightarrow PO_4^{3-} + 2H^+$$
 (16)

with pKa's of 7.21 and 12.63, whereas for the borate/boric acid system the buffering reactions are:

$$H_3BO_3 + OH^- = B(OH)_4^-$$
 (17a)

$$2H_3BO_3 + OH^T = H_7B_2O_7^T$$
 (17b)

$$3 H_3 BO_3 + OH^- = H_{10} B_3 O_{10}^-$$
 (17c)

and the pKa's are 9.14, 12.74 and 13.80.

To determine the appropriate amount of the buffer to be stored for controlling the pH of the sump water, the buffer capacity near the desired pH must be large. The buffer must be able to preserve a high pH despite the overall H⁺ production expected from the radiolytic production of inorganic and organic acid. Note that the initial pH of the sump water is also important to consider and additional base may be required to bring the pH up to a region where the buffer can act effectively. For example, although boric acid is an ideal buffer and is already present in coolant in many types of reactors, its effectiveness as a buffer will be dependent upon the pH at which the coolant will be introduced into containment. If the coolant is maintained near pH 7 during plant operations, additional base will be required to bring the pH up to a point that the borate can act as an effective buffer under accident conditions.

It should be noted that these candidate buffers are hygroscopic and could cake up during long-term storage in contact with moist air. Hence, a delivery system that ensures dissolution of buffering agent by water in the event of an accident is a requirement for a mitigation system.

2.3 Iodine behaviour in the gas phase

The aqueous chemistry of iodine offers at least two mechanisms for management of the iodine source term in a reactor accident – adsorption of iodine on submerged surfaces such as submerged silver surfaces and control of the solution pH. Another approach toward managing the iodine source term is to enhance the removal of iodine suspended in the containment atmosphere. A management strategy addressing the airborne concentration of iodine might be adopted in preference to or as a supplement to a strategy focused on the aqueous chemistry of iodine. Design of an accident management strategy focused on the atmospheric concentration of iodine requires some appreciation of the chemical and physical processes affecting iodine suspended in the containment atmosphere. The important chemical and physical processes are discussed in this section of the chapter.

Gaseous iodine may be injected into the containment atmosphere as a result of the processes leading to release of iodine from fuel and the reactor coolant system or as a result of partitioning of volatile forms of iodine from water. It may also be possible to form gaseous iodine species from iodine-bearing aerosols as a result of hydrogen burns in containment atmospheres that are not inerted. Hydrogen combustion events in the containment can produce local, transient, temperatures that are sufficiently high that metal iodides thermally decompose to release atomic iodine or hydrogen iodide (HI). Understanding is yet incomplete on the details and, especially, the efficiency of the decomposition of iodine-bearing aerosols in hydrogen combustion events. There is some evidence that steam inhibits the formation of molecular iodine. Also, other aerosol materials that are reactive toward high-temperature cesium hydroxide may accentuate the release of iodine species during combustion events.

Hydrogen combustion could be an episodic, repeated event in the progression of an accident. At the current state of understanding of the effects of hydrogen combustion events, it is prudent for the planning of source term management to assume some transient increases in the gaseous iodine concentration in containment will be associated with combustion events. These increases in the concentration of gaseous iodine in the containment atmosphere as a result of combustion events would be expected to decay away as a result of other processes discussed in this section. An interesting and unresolved question is whether active or passive hydrogen recombiners can cause significant decomposition of iodine-bearing aerosols and cause steady-state increases in the concentrations of gaseous iodine in the containment.

The chemical forms of gaseous iodine in the containment atmosphere that are of interest are molecular iodine and organic iodides whether these species reached the containment as a result of iodine release from the fuel, partitioning of volatile iodine species from water or as a result of combustion processes. Another volatile form of iodine, HI, is very hygroscopic and will rapidly dissociate in contact with water to form solvated Γ . There can be some conversion of molecular iodine, $I_2(g)$, into organic iodides, RI, in the radiation field of the containment atmosphere:

$$RH(g) + \bullet OH(g) \rightarrow R\bullet(g) + H_2O$$
 (21)

$$R^{\bullet}(g) + I_2(g) \rightarrow RI(g) + I^{\bullet}(g)$$
 (22)

The key reactant, the hydroxyl radical, OH, is formed by the radiolysis of water vapour. Concentrations of this key reactant are much lower in the gas phase than in the aqueous phase because of the higher density and higher radiation adsorption of water. Consequently, gas phase formation of organic iodides from molecular iodine is not usually an important process relative to the formation of organic iodides in the aqueous phase and partitioning of these iodides into the atmosphere.

Both molecular iodine and organic iodides are susceptible to thermal and radiolytic decomposition in the containment atmosphere. Of particular interest is the reaction of iodine species with ozone formed by radiolytic processes in the atmosphere:

$$I_2(g) + O_3 \rightarrow I_xO_y$$
 (23)

The iodine-bearing products of reaction are solid and not volatile. That is, iodine-bearing aerosols are formed by the reaction. These iodine aerosols are subject to all the deposition processes discussed in connection with other aerosols in Chapter V. There has been a proposal to enhance gaseous iodine removal by augmenting ozone production in the containment atmosphere using ultraviolet photolysis of oxygen. The concentration of ozone that would be needed to make a significant reduction in the airborne iodine concentration is large due to the slow rates of homogeneous, gas phase reactions. Ultraviolet absorption by other aerosols may also limit the effectiveness of the technique.

More important than the homogenous, gas phase reactions of iodine species are the reactions of these species with surfaces and the physical removal of the species by containment sprays. Safety analyses often credit the gaseous iodine attenuation by containment sprays. The main purpose of the containment sprays is to reduce the pressure in containment by condensing steam. Fresh spray droplets generated when the sprays are first activated are also quite effective in removing gaseous molecular iodine. The capability of spray droplets to remove gaseous iodine can be enhanced with additives to the spray to raise the pH (such as NaOH) or to reduce molecular iodine to iodide ion (such as Na₂S₂O₃). Later in an accident, when sprays are operated in the recirculation mode drawing water from the contaminated sumps where volatile iodine is being generated, there will be no attenuation of the gaseous iodine concentration. On the contrary, spraying can enhance the mass transport of volatile iodine from the aqueous phase to gas phase and increase the gaseous iodine concentration in the containment atmosphere.

The adsorption of gaseous iodine species on surfaces has received much attention in the reactor safety literature. Varieties of metal, concrete and painted surfaces are in a reactor containment building. The gaseous forms of iodine will interact with these surfaces. The interactions are usually thought to involve a rapid physical adsorption followed by a slower chemical reaction. Dry (or moist, but not wet) surfaces of both stainless steel and organic (vinyl-, epoxy-, polyurethane-) paint have large capacities for absorbing molecular iodine. The adsorption rate of molecular iodine on the dry surfaces is close to the limit dictated by gas-phase mass transport. Intermediate-scale studies performed in the Radioiodine Test Facility have shown that about 75 to 95% of airborne molecular

iodine can be immobilised, at least temporarily, on dry surfaces. It is to be cautioned, however, that these results are only indicative of the importance of surface adsorption of gaseous iodine species. The fraction of gaseous iodine adsorbed on surfaces in a reactor containment building will differ from that observed in test facilities because of the much lower surface to volume ratio and the different mix of surface materials.

There have been some efforts to develop paint with a high adsorption capacity for iodine (the so-called 'reactive' organic coating). When dry, such a coating could augment iodine removal. But, surfaces in reactor containment will be wet during major portions of a reactor accident as a result of steam condensation and the actions of sprays. Gaseous iodine species interacting with such wet surfaces will undergo aqueous chemistry similar that discussed above. The difference is, of course, that it is much more difficult to control the pH of water films on surfaces in the containment than it is to control the pH of bulk water bodies in the containment. Eventually, water films will drain into the larger bodies of water where pH can be controlled.

VI.3 Containment iodine codes

The chemical and physical processes that affect iodine under reactor accident conditions are quite varied and have only been synoptically described in this chapter. Fortunately, a number of computer codes have been developed to provide more detailed descriptions of these processes. The computer codes IMPAIR, IODE, TRENDS, MELCOR, INSPECT, LIRIC and IMOD, have been used in assessing the safety of nuclear installations. Although they still require improvements, many of these codes have been applied to simulate various intermediate- and bench-scale results with some success. A significant code comparison and validation exercise has been the International Standard Problem (ISP)-41: Computer Code Exercise Based on a Radioiodine Test Facility (RTF) Experiment on Iodine Behaviour in Containment under Severe Accident Conditions. Although the conditions of the test chosen for this exercise were far different than the conditions expected during real accidents, the results from this exercise provide a good understanding of the limitations of each code. The results have helped to identify the areas of improvement needed to increase confidence in the validity and accuracy of these tools for assessing the safety of nuclear installations.

There have been two different approaches to model development for predicting iodine behaviour for conditions expected to be typical of an accident: empirical (IMPAIR, IODE, TRENDS, IMOD) and mechanistic (INSPECT, LIRIC). The ISP exercise established that both the mechanistic and empirical modelling approaches are reasonable since all of the models were able to reproduce the test results. Currently, there are only a few tests that can be used to validate the models over a wide range of conditions. As a result, the applicability of most of the codes is limited and the uncertainty limits are not well defined.

VI.4 Removal of gaseous iodine by filters

Pathways for venting the containment atmosphere may be provided for a number of reasons, and these pathways may be equipped with filters to remove iodine from the vented gas. Filters for iodine removal can be present in both passive systems (in which flow continues only as long as there is a pressure difference) and active systems (in which there is a continuous forced flow at a controllable rate). Dry filters intended for the removal of aerosol particles are not likely to be effective for the removal of gaseous forms of iodine, especially organic iodides. Even if gaseous iodine will absorb on the filter medium, heat loads on the filter medium caused by radioactive decay can lead to revaporization of the absorbed iodine. Filters that involve water must be maintained at high pH to avoid the formation of volatile forms of iodine by processes identical to those that occur in reactor containment sumps.

Filters specific to the retention of gaseous forms of iodine are made with charcoal. Charcoal used in these filters is highly activated (i.e., surface area of the order of 1000 m²/kg). Usually, the charcoal filters are also impregnated with TEDA (tri-ethylene-di-amine) to increase trapping efficiency for organic iodides (TEDA reacts with organic iodides to form quaternary ammonium salts). A large body of work has been carried out in support of filters used in active ventilation systems. This work has shown that TEDA charcoal is a very effective means of removing various species of volatile iodine from a gas flow and retaining them. For use of such filters over a prolonged period, measures are needed to ensure that the filter medium retains the characteristics necessary for reliable trapping and retention of iodine species from the gas flow. The most important of these are measures to control the relative humidity of air entering the charcoal, and to limit the temperature of the charcoal. Humidity control is important because high humidity significantly reduces the adsorption capacity and causes condensation on the charcoal, reducing the effectiveness of charcoal enormously. Temperatures rise in a charcoal filter because of both the heat of adsorption and because of the decay heat from fission products trapped in the filter. Temperature control is important because iodine desorbs more readily at higher temperatures. The major concern regarding temperature, however, is the low ignition temperature of TEDA vapours (the flash point is about 180 °C).

The effectiveness of active filter systems is known to be high, based on system tests, research results and simulations. The effectiveness of filters in passive systems as a means of limiting the potential for iodine releases is not known. Backfitting a filter system to an existing containment may be more expensive and less effective than some combination of other mitigation techniques discussed below. A drawback for all charcoal filter systems is the high cost associated with periodic testing of the effectiveness of charcoal.

VI.5 Summary of potential mitigation methodologies

The technical discussions of the chemical and physical behaviour of iodine in containment presented in the previous sections of this chapter give a summary of the key processes and factors that can influence the airborne iodine concentration during a severe accident. Based on this material, six options that could be used to limit the airborne iodine concentration are outlined below.

(1) Maintain the pH of the sump water at a high value

The concentration of volatile iodine species in solution decreases by about an order of magnitude with an increase in pH by one unit over the pH range of ~5 to ~10. The concentration of airborne iodine during an accident can therefore be kept low by maintaining the pH high (about 9 is considered adequate). There are several factors that need to be considered in assessing the requirements for a chemical buffer to be added to containment sump water in the event of a severe accident to maintain such a pH. These factors include estimates for the rate of acid production and the stability and solubility of the buffering agent.

(2) Zinc-Primer-coated surfaces

There is evidence that surface coatings containing zinc metal can adsorb and retain iodine from water at a high pH (9 -10). Continued retention of the iodine requires that the water in contact with the zinc coating be maintained at a high pH. Hence, for this mitigation strategy to be effective, it must be combined with the use of pH buffer to control the pH of the water in containment.

(3) Silver immersed in the sump water

Silver in contact with water is a very effective iodine adsorber. Silver in a form having a high specific surface area (e.g., silver wool) could be located in the containment sump to react with and immobilise iodine in the sump water. This mitigation option is conceptually similar to the use of zinc in a surface coating. It is, however, more expensive due to the value of the silver. In addition, design steps would have to be taken to ensure that the silver was not subject to long-term degradation through reaction with atmospheric impurities, and particularly sulfides, which would compromise its effectiveness.

(4) Ozone production by UV photolysis

The reaction of molecular iodine with ozone in the gas phase leads to the production of non-volatile iodine oxides that subsequently dissolve in water to form non-volatile iodate ions. Ozone production using UV photolysis of oxygen in the containment air would increase the rate of formation of iodine oxides. The concentration of ozone that would be required to affect significantly the airborne iodine concentration is quite large because of the slow rates of homogeneous, gas phase reactions. During an accident, the containment atmosphere may contain aerosols that would absorb UV light, inhibiting ozone production. This mitigation option may have limited effectiveness.

(5) Paint with a high adsorption capacity for iodine

There have been some efforts to develop paint with a high adsorption capacity for iodine. However, such paint may not help much in terms of the overall iodine sorption during an accident. The overall adsorption rate on dry surfaces under accident conditions is limited by the gas phase mass transport. Furthermore, because water films may cover surfaces, there appears to be limited scope for using surface reactions to immobilise iodine and reduce the gas phase concentrations.

(6) Filtered venting

Systems designed to vent the containment atmosphere and to filter molecular iodine from the vented flow are in service and a body of research results characterizing the effectiveness of these systems also exists. Application of the research data to other systems of different design is possible in principle. Retention capability of wet scrubbers for organic iodides are not well established.

VI.6 Accident management as practised in OECD Member countries

The PWG4 Task Group on Containment Aspects of Severe Accident Management (CAM) decided in its March 1998 meeting to collect information from the Member countries on specific features of currently operating nuclear power plants. This information was collected to compile a status report on the current practices, implemented engineered and passive systems for the accident management aspects of the control of the release of iodine, cesium, strontium and other fission products in the containment during a severe accident. The intention is to couple the currently available systems and practices with the state of the art information provided by the scientific experts.

A questionnaire was prepared to collect the information from the Member countries on the current practices on the management of gaseous iodine during a postulated severe accident. This section compiles the information provided in responses to the questionnaire.

The currently operating plants were licensed to cope with the conditions expected during postulated design basis accidents. Some of the plants were equipped from the beginning or were backfitted with dedicated systems which are effective at removing gaseous iodine normally for design basis accidents when some limited gap release of iodine might be expected. HEPA filters are included

in some plant designs, for example. Since these systems were not designed to cope with the severe conditions postulated during a core melt down accidents, accident management of iodine removal including engineered systems in addition to the natural depletion processes have become important. The processes of formation and destruction of gaseous iodine can persist for many days. Consequently, gaseous iodine can be a safety issue for a long time after an accident starts. Long term formation and leakage of gaseous iodine is a particular threat to the safety of site operators.

6.1 Practices and applications adopted by the Member countries

Table 1 presents the list of Member countries that returned the questionnaire. Appendix A at the end of the report presents the information that was provided. Attempts to manage airborne iodine in the containment and its controlled release into the environment vary between two extremes. One extreme is that in some power plants only natural depletion mechanisms are relied upon to mitigate the airborne iodine concentration in the containment and no other active or passive measures are currently foreseen. At the other extreme are engineered measures to control the pH and to fix the iodine in water. This is often done with additives to the spray or sump water and by controlling the venting to the environment with a containment venting filter or plant emergency filtration system. Tables 2 to 7 indicate the range of practices reported in each country. Since the information provided does not cover completely all the operating power plants in the Member countries, it is not practical to make statistics on practices and applications. The following sections overview various aspects of engineered measures.

6.2 Spray systems

The main function of a spray system in a power plant is to condense the steam generated during a LOCA or steam generated if some of the debris, leaving the reactor pressure vessel, boils the sump and if the sump cooling fails. Sprays not only condense the steam but also effectively remove aerosol particles and some of the gaseous molecular iodine provided that spray droplets act as a sink and do not become saturated with volatile iodine. Droplets may reach saturation during their fall or may become saturated during the recirculation mode of spray operation. This is especially so if spray additives lose their effectiveness due to thermal and radiolytic decomposition. When saturated, the droplets behave as additional iodine sources in the containment atmosphere. In order to avoid iodine release from the spray droplets back into the atmosphere, the additives summarised in Table 2 are used to trap the iodine in the droplets by converting it into a non-volatile form.

6.3 Containment sump additives

Most initial efforts to develop accident management techniques have focused on maintaining high pH in water within the containment. Some utilities take advantage of adding additives to spray and hence increasing the sump water pH to a high value. A few utilities have deposited bags of trisodium phosphate in the reactor containment. Other utilities use hydroxides. Table 3 presents the current practices varying from no pH control to the use of additives.

6.4 Containment filters

Containments are normally equipped with HEPA and activated charcoal filters to continuously clean the containment atmosphere during power operation and shutdown periods. These are designed for dry airflow containing particulate and iodine. Severe conditions, such as high temperatures, large steam flows, condensing steam, high aerosol loads and high iodine loads, that will exist in a containment during a severe accident, may reduce the efficiencies of the filters if additional engineering measures have not been provided to minimize the adverse effects. Tables 4 and 5 present the filters and containment venting filters in use currently and also the decontamination factors expected of these filters. Some utilities may use the filters with additional equipment upstream such as the containment atmosphere emergency filtration system.

6.5 Passive iodine removal

The purpose of this question is to find out whether any passive sink for the iodine has been provided to supplement the natural iodine removal mechanisms, like deposition, adsorption, chemical reaction, mass transfer into the water pool or into the droplets, and pool scrubbing, etc. Table 6 presents a summary of the responses. Except the borax used in the ice condenser of the Loviisa units (Finland) no other passive means have been reported.

6.6 Iodine mitigation measures implemented/planned

The purpose of this question was to find out whether any additional iodine mitigation measures have been planned or already implemented other than using additives in the spray and containment sump water and using controlled containment venting with a filter. Other than these measures, if already implemented, there are no other mitigation measures reported by the participating organizations.

6.7 Measurement of iodine in containment atmosphere and controlled iodine release into the environment

Activity levels in the containment are determined. However, as shown in Table 7, some of the plants have devoted further efforts to measure molecular and organic iodine speciation. The same is also true for the controlled release of iodine into the environment.

6.8 Other accident management measures to mitigate airborne iodine

No participating organisation responded to this question.

6.9 Conclusions

The most common technique adopted at existing nuclear power plants to suppress iodine volatility is to maintain high values of pH in bodies of water within reactor containment. High pH converts molecular iodine into a nonvolatile form and suppresses formation of organic iodides. Establishing high values of pH throughout the various water bodies in reactor containment and maintaining the high values of pH over time impose design requirements and requirements to manage pH following accidents. Spray systems are being used to suppress release of airborne iodine from the "breathing" of paints on containment surfaces. Sprays will do this effectively as long as the spray water has sufficiently low concentrations of volatile forms of iodine. In general, this constraint on the effectiveness of sprays has not been considered in the design of accident management strategies. Therefore, maintaining containment integrity as long as possible or providing for controlled releases of iodine with some filtration or other mechanism to reduce iodine concentrations seem to be the most effective ways to mitigate the long term releases of iodine into the environment.

Table 1: List of countries that returned the questionnaire (Status: April 1999)

Belgium	Canada
Czech Republic	Japan
Spain	The Netherlands
Finland	Korea
USA	France
Sweden	Switzerland
Germany	

Table 2: Spray System Characteristics

Additives	Korea	Finland	Spain	Belgium	Sweden	France (1)	Czech Rep.	Canada	The Netherlands	The USA (1)	Japan	Switzerland	Germany
NaOH	25 - 32 %		30 %	20-30%									
КОН		Max 423 mg/kg											
LiOH					2								
Trisodium phosphate					2								
Hydrazine	30-35 % 50-100 mg/kg	Max 106 mg/kg					0.2gN ₂ H ₄ . H ₂ O+2.7g KOH +7gH ₃ BO ₃ per liter of spray solution	100 mg/kg			35 %		
Boric acid				*			*						*
No additives	*		*					*	*		*	*	*

¹⁾ No information provided

²⁾ Additives used but the concentration was not provided

Table 3: Containment sump conditions

Additives	Korea	Finland	Spain	Belgium	Sweden	France	Czech Rep.	Canada	The Netherlands	The USA	Japan	Switzerland	Germany
No pH control	*		*	*			*	*	*	*	*	*	*
NaOH				pH: 7-10 Recirculation phase						pH: >7	pH: 7 (2)		
Trisodium phosphate	pH: 7- 10		pH: 7.2							pH: >7			
LiOH					pH: 8-8.5 (1)								
Hydrazine													
КОН		pH: 8 - 9											
Na ₂ O(B ₂ O ₃)													
B(OH) 3													
Boric acid+Soda						pH: high							

¹⁾ Information is provided only for BWR, ? for PWRs $\,$

Added through containment spray line

Containment filters and decontamination factors Table 4:

	Korea	Finland	Spain	Belgium	Sweden	France	Czech Rep.	Canada	The Netherlands	The USA	Japan	Switzerland	Germany
none	*									*		(2)	(3)
НЕРА	99.95- 99.99%	min 99.75%	99.97% DOP	99%	(1)	(1)	DF: 1000 - 10000	the exhaust line of normal filtered air discharge system		95-99%	>99%		(3)
Active carbon	95%-99%	99% CH₃I 99.9% I₂	95-99%	(KI) >90% I _{2,} & CH ₃ I	(1)		DF: 10 for I ₂	the exhaust line of normal filtered air discharge system	(1)	95-99%	95%- 99.99%		(3)
Prefilter (aerosol only)				85-95 %	(1)			the exhaust line of normal filtered air discharge system		95-99%	(1)		(3)

Efficiencies are not provided
 No special filters for severe accidents
 No information is provided

Table 5: Controlled Containment Venting and decontamination factors

	Korea	Finland	Spain	Belgium	Sweden	France	Czech Rep.	Canada	The Netherlands	The USA	Japan	Switzerland	Germany
none	*	*	*	*			*			*	*		
wetwell pool scrubbing											Suppress. pool scrubbing (1)		
venting filter					multiventuri system DF> 100- 500				Venturi scrubber and metal filter I ₂ : 99.5% CH ₃ I: >90%			(2) DF: I: 100 min	(3)
internal metallic prefilter+ sand-bed filter						*							
Emergency Filtered Air Discharge System								two trains of filters, containing demisters, heaters, prefilters, two HEPA filters and carbon filters					
carbon bed	99.5 %												

¹⁾ NPP Kashiwazaki Kariwa -3 may vent directly by venting the wetwell gas space into the environment without involving additional filter in between.

²⁾ Sulzer-wet aerosol scrubber; additives: sodium carbonate and sodium thiosulfate; Multi Venturi scrubber; Venturi scrubber with integrated metal mesh filter

³⁾ Venturi scrubber, metal fiber, molecular sieve, capacity: PWR: 60 kg, BWR: 30 kg, efficiency: aerosol: 99.9 %, elem. Iodine: 90 %

Table 6: Passive iodine removal

	Korea	Finland	Spain	Belgium	Sweden	France	Czech Rep.	Canada	The Netherlands	The USA	Japan	Switzerland	Germany
Natural (1) processes	*	*	*	*	*	*	*	*	*	*	*	*	*
Special means		Borax in ice condenser traps efficiently the I ₂									standby gas treatment system		

¹⁾ Adsorption/deposition on walls and aerosol particles, removal by mass transfer into pool water or into droplets, wetwell pool scrubbing, etc.

Table 7 Measurement of iodine in containment atmosphere and controlled iodine release into the environment

	Korea	Finland	Spain	Belgium	Sweden	France	Czech Rep.	Canada	The Netherlands	The USA	Japan	Switzerland	Germany
In Containment	post- accident air sampling and on-line radiation monitoring	with speciation	*	with speciation	global dose rate	global dose rate	gaseous iodine	iodine activity	*	iodine activity		Yes (1)	Yes (2)
Into Environment	on-line radiation monitoring	with speciation	*	with speciation	*	*	None	iodine activity	*	iodine activity	*	Yes (1)	

Detailed information not provided
 Sampling system in preparation to be installed mainly for PWRs

VI.7 Annotated bibliography of iodine chemistry in reactor accidents

- 1. **Proceedings of the First CSNI Workshop on Iodine Chemistry in Reactor Safety**, A.M. Deane and P.E. Potter, editors, AERE-R-11974, United Kingdom Atomic Energy Authorities, Oxfordshire, United Kingdom, January 1986.
- 2. **Proceedings of the Second CSNI Workshop on Iodine Chemistry in Reactor Safety**, A.C. Vikis, editor, AECL-9923, CSNI Report 149, Atomic Energy of Canada, Ltd., Toronto, Canada, March 1989.
- 3. **Proceedings of the Third CSNI Workshop on Iodine Chemistry in Reactor Safety**, K. Ishigure, M. Saeki, K. Soda, and J. Sugimoto, editors, JAERI-M-92-012, NEA/CSNI/R(91)15, Japan Atomic Energy Research Institute, Tokai-mura, Naka-gun, Ibaraki-ken, 319-11, Japan, 1992.
- 4. **Proceedings of the Fourth CSNI Workshop on the Chemistry of Iodine in Reactor Safety**, S. Güntay, editor, PSI Report Nr. 97-02, NEA/CSNI/R(96)6, Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland, December 1996.

The proceedings of these four conferences provide a good indication of the issues and the current understanding of iodine partitioning from water pools into the containment atmosphere. Excellent papers in these conference proceedings address topics considered in this chapter and addressed in this bibliography.

- 5. J.C. Wren, G.A. Glowa, and J.M. Ball, "Modeling Iodine Behaviour Using LIRIC 3.0", In **Proceedings of the Fourth CSNI Workshop on the Chemistry of Iodine in Reactor Safety**, Würenlingen, Switzerland, 1996, p. 507 (1997).
- 6. S. Dickinson and H.E. Sims, "Modifications to the INSPECT Model", In **Proceedings of the Fourth CSNI Workshop on the Chemistry of Iodine in Reactor Safety**, Würenlingen, Switzerland, 1996, p. 487 (1997).
- 7. M. Furrer, R.C. Cripps, and E. Frick, **Iodine Severe Accident Behavior Code IMPAIR 2**, PSI Bericht Nr. 25, Paul Scherrer Institut, SH-5232 Villigen/PSI, Switzerland.
- 8. C. Hueber, S. Dickinson, and M. Pignard, Validation du code IODE: Modélisation des réactions d'hydrolyse et de Dushman, N.T. IPSN/SEMAR no. 93/45, 1993.
- 9. Weber, C.F., Kress, T.S., Beahm, E.C., Shockley, W.E., Darsh, S.R., "TRENDS: A Code for Modelling Iodine Behaviour in Containment During Severe Accidents", In **Proceedings of the International Cent. Heat Mass Transfer**, **30**, 665-674 (1990).
- 10. J.M. Ball, et Al., "INTERNATIONAL STANDARD PROBLEM (ISP) NO. 41: Computer Code Exercise Based on a Radioiodine Test Facility (RTF) Experiment on Iodine Behaviour in Containment under Severe Accident Conditions", In **Proceedings of OECD Workshop on Iodine Aspects of Severe Accident Management**, NEA/CSNI/R(99)7, Vantaa, Finland, 1999.
- 11. J.C. Wren, G. Glowa and J.M. Ball, "A Simplified Iodine Chemistry and Transportation Model: Model Description and Some Validation Calculations", In **Proceedings of OECD Workshop on Iodine Aspects of Severe Accident Management**, NEA/CSNI/R(99)7, Vantaa, Finland, 1999.

References 5 - 11 describe containment iodine codes that are used in assessing the safety of nuclear installations.

12. J.C. Wren, J.M. Ball and G.A. Glowa, "The Interaction of Iodine with Organic Material in Containment", **Nuclear Technology**, **125**, 337, 1999 March (1999).

- 13. R. Fluke, J.M. Ball, S. Dickinson, H.E. Sims and G.J. Evans, **Iodine Behaviour in Containment. Quantifying the Effects of Organic Reactions**, EPRI, ACEX TR-B-04, (1997).
- 14. E.C. Beahm, Y.M. Want, S.J. Wisbey, W.E. Shockley, "Organic Iodide Formation During Severe Accidents in Light Water Nuclear Reactors", **Nuclear Technology**, **78**, 34, (1987).
- 15. J. Wing, **Post-Accident Gas Generation from Radiolysis of Organic Materials**, United States Nuclear Regulatory Commission Report, NUREG-1081, (1984).
- 16. A. M. Deane, **Organic Iodine Chemistry Relevant to Nuclear Reactors: A Review**, AERE-R-12359, Harwell Laboratory, Oxfordshire, United Kingdom, March 1988.
- 17. Postma, A.K. and Zavadoski, R.W., **Review of Organic Iodide Formation Under Reactor Accident Conditions in Water-Cooled Reactors**, Atomic Energy Commission Report, WASH-1233, (1972).

References 12-17 provide background reading on the effects of organic reactions on iodine volatility.

- 18. J.M. Ball, W.C.H. Kupferschmidt and J.C. Wren, "Phase 2 Radioiodine Test Facility Experimental Program", In **Proceedings of the Fourth CSNI Workshop on the Chemistry of Iodine in Reactor Safety**, Würenlingen, Switzerland, 1996, p. 63 (1997).
- W.C.H. Kupferschmidt, J.B. Buttazoni, A.J. Melnyk, R. Portman and G.G. Sanipelli, "The Effects of Zinc Primer Surfaces on Iodine Behaviour Within Containment", In Proceedings of the Third CSNI Workshop on Iodine Chemistry in Reactor Safety, Toki-Mura (Japan), 1991, Japan Atomic Energy Research Institute Report JAERI - M 92-012, p. 195 (1992).
- 20. W.C.H. Kupferschmidt, J.M. Ball, J.B. Buttazoni, G.J. Evans, A.J. Melnyk, A.S. Palson, R. Portman and G.G. Sanipelli, **Final Report on the ACE/RTF Experiments**, ACE-TR-B3, (1992).
- 21. D. Jacquemain, N. Hanniet, **An Overview of the Gaseous Iodine Data in the Experimental Circuits and the Containment in the first two PHEBUS Tests FPT-0 and FPT-1**, Document PHEBUS PF n° IP/99/428 (1999)

References 18-21 present intermediate- and large-scale test results.

22. C B. Ashmore and H.E. Sims, **Nitric acid formation in irradiated moist air**, AEAT-0570, AEA Technology, Winfrith, Dorset, United Kingdom, October 1996.

This paper provides a thorough review of the processes and the issues associated with the formation of nitric acid by the irradiation of moist air as would be expected in reactor containment during accidents.

- 23. S.B. Srivastava, K.V.S. Rama Rao, L.V. Shastry and A.S. Sarpotdar, **Compatibility Studies on Vinyl Paint**, Government of India, Atomic Energy Commission Report, BARC-322, Bhabha Atomic Research Centre, Bombay, India, (1967)
- 24. W.C.H. Kupferschmidt, J.B. Buttazoni, G.J. Evans, B.L. Ford, A.S. Palson, R. Portman and G.G. Sanipelli, "Iodine Chemistry in the Presence of Vinyl Surfaces", In **Proceedings of the Third CSNI Workshop on Iodine Chemistry in Reactor Safety**, Toki-Mura (Japan), 1991, Japan Atomic Energy Research Institute Report JAERI M 92-012, p. 213 (1992).
- 25. A.C. Vikis, **The Photochemical Method for Radioiodine Abatement**, Atomic Energy of Canada, Ltd. Report, AECL-7819, 1984.

Reference 25 proposes to augment the ozone production from the radiation field in a reactor containment by photochemical means.

- 26. J.C. Wren, G.A. Glowa and J. Merritt, "Corrosion of Stainless Steel by Gaseous Iodine", J. Nuclear Materials, 265, 161-177, (1999).
- 27. H.S. Rosenberg, J.M. Genco and D.L. Morrison, Fission Product Retention and its Enhancement under Reactor Accident Conditions: Deposition on Containment-System Surfaces, BMI-1865, (1969).
- 28. S. Hellmann, F. Funke, G-U. Greger, A. Bleier and W. Morell. "The Reaction Between Iodine and Organic Coatings Under Severe PWR Accident Conditions An Experimental Parameter Study", In **Proceedings of the Fourth CSNI Workshop on the Chemistry of Iodine in Reactor Safety**, Würenlingen, Switzerland, 1996, p. 345 (1997).
- 29. A. Zoulalian and E. Belval-Haltier, "Interaction between Molecular Iodine in Gaseous Phase and a Coat of Paint Influence of Temperature, Humidity, and Hydrothermal Treatment on Iodine Trapping Kinetics", Nuclear Technology, 122, 196 (1998).
- 30. H.S. Rosenberg, G.E. Cremeans, J.M. Genco, D.A. Berry and D.L. Morrison, **Fission-Product Deposition And Its Enhancement Under Reactor Accident Conditions: Development Of Reactive Coatings**, Battelle Memorial Institute Report, BMI-1874, Columbus, OH., (1969).
- 31. H.E. Sims, S. Dickinson, G. Evans, J.C. Wren, G. Glowa, **Iodine Behaviour in Containment: The Reaction of Iodine with Surfaces**, EPRI ACEX TR-B-06 (1997)

References 26-31 provide background information on the surface interaction of gaseous iodine.

- 32. F. Funke, G.-U. Greger, A. Bleier, S. Hellmann and W. Morell, "The Reaction Between Iodine and Silver Under Severe PWR Accident", In **Proceedings of the Fourth CSNI Workshop on the Chemistry of Iodine in Reactor Safety**, Würenlingen, Switzerland, 1996, p. 283, (1997).
- 33. E. Krausmann, Y. Drossinos, "A model of silver-iodine reactions in a light water reactor containment sump under severe accident conditions", **J. Nuclear Materials**, **264**, 113, (1999).
- 34. N. Ishiwatari, "Aqueous Solution of Iodide Under Gamma Ray Irradiation Fixation of iodide by adding soluble silver salt", In **Proceedings of the Third CSNI Workshop on Iodine Chemistry in Reactor Safety**, JAERI-M 92-012, 1991, Japan Atomic Energy Research Institute Report JAERI M 92-012, p. 81 (1992).
- 35. R. Cripps, S. Guentay, H. Bruchertseifer, **Investigation of Stability of AgI in Form of Colloids in an Electron Beam**, CEC ST-ICP(97)19, (1998).

References 32-35 deal with silver-iodine reactions.

- 36. J.C. Wren, W. Long, C.J. Moore and K.R. Weaver, "Modelling the Removal and Retention of Radioiodine by TEDA-Impregnated Charcoal under Reactor Accident Conditions", **Nuclear Technology**, **125**, p. 13 (1998)
- 37. V.R. Deitz and L.A. Jonas, "Catalytic Trapping of Methyl Iodide by Beds of Impregnated Charcoal", **Nuclear Technology**, **37**, 59 (1978).
- 38. H. Shiomi, Y. Yuasa, A. Tani, M. Ohki and T. Nakagawa, "Parametric Study of Removal Efficiency of Impregnated Activated Charcoal and Silver Zeolite for Radioactive Methyl Iodide", **Proceedings of 17th DOE Nuclear Air Cleaning Conf.**, Denver, Colorado, August 2-5, 1982, CONF-820833, p. 199, U.S. Department of Energy (1983).

- 39. H. Deuber, K. Gerlach, V. Giraud and R. Kaempffer, "Investigations on the Aging of Various Activated Carbons in the Exhaust Air of a PWR over an Extended Period of Time", **Proceedings of 19th DOE/NRC Nuclear Air Cleaning Conf.**, Seattle, Washington, August 18-21, 1986, CONF-860820, p. 243, U.S. Department of Energy (1987).
- 40. B.H.M. Billinge and J.B. Doeherty and M.J. Bevan, "The Desorption of Chemisorbed Oxygen From Activated Carbons and Its Relationship to Aging Methyl-Iodide Retention Efficiency", **Carbon**, **22**, 83 (1984).
- 41. ASTM Standards, D3803-79, "Standard Test Methods for Radioiodine Testing of Nuclear-Grade Gas-Phase Adsorbents", The American Society for Testing and Materials, Philadelphia, Pa. (1980).

Reference 36 - 41 provides experimental and modelling studies on the effectiveness of charcoal filters for removing iodine from gaseous stream.

42. L.E. Herranz and F. Robledo, "A Potential Strategies to Control Iodine Released into the Containment in the Case of a Severe Reactor Accident", In the Proceedings of **OECD Specialist Meeting on Selected Containment Severe Accident Management Strategies**, NEA/CSNI/R(95)3, Stockholm, June 1994.

A comprehensive discussion is presented in reference 42 of the various methods now being considered to attenuate the amounts of gaseous iodine in reactor containment.

- 43. M.L. Hyder, "Measurements of Iodine Uptake in Falling Water Droplets", **Nuclear Technology**, **94**(1991)80.
- 44. M.F. Albert, J.S. Watson, and R.P. Wichner, "The Absorption of Gaseous Iodine by Water Droplets", **Nuclear Technology**, **77**(1987)161.
- 45. B.A. Soldano and W.T. Ward, "A Study of the Absorption of CH₃I by Aqueous Spray Solutions", **Nuclear Technology**, **10**(1971)460.
- 46. Y. Nishizawa, S. Oshima, and T.Maekawa, "Removal of iodine from atmosphere by sprays", **Nuclear Technology**, **10**(1971)486.
- 47. R.K. Hilliard, A.K. Postma, J.D. McCormack, and L.F. Coleman, "Removal of Iodine and Particles by Sprays in the Containment Systems Experiment", **Nuclear Technology**, **10**(1971)499.

Removal of gaseous iodine from the containment atmosphere by sprays is a classic reactor safety measure. These papers describe the current state of understanding of the removal process and expected efficiencies of spray removal of molecular iodine and organic iodides.

- 48. M.L. Brown, E.C. Beahm, and W.E. Shockley, **The Oxidation of Cesium Iodide in Stationary Pre-mixed Hydrogen Flames**, ACE-TR-B6, ORNL/M-1080, Oak Ridge National Laboratory, Oak Ridge, TN, 1990.
- 49. W.C.H. Kupferschmidt, et al., "The Oxidation of Cesium Iodide During Hydrogen Combustion", In Proceedings of the Third CSNI Workshop on Iodine Chemistry in Reactor Safety, Toki-Mura (Japan), 1991, Japan Atomic Energy Research Institute Report JAERI M 92-012, p. 298 (1992).

These papers describe the effects of hydrogen combustion on the stability of CsI and the possible formation of molecular iodine by oxidation of CsI. The combustion problem is complicated and formation of molecular iodine may be inhibited by the presence of high concentrations of steam.

VII. MITIGATION OF RELEASE FROM THE CONTAINMENT

Mitigation of the release of radionuclides from reactor containments once these containments are open to the environment has not received a great deal of attention. In considering possible accident management steps in this area it is useful to distinguish between accidents initiated during power operation when the containment is intact and accidents initiated during shutdown operations when the containment is initially open to the environment.

VII.1 Mitigation of At-power accidents

Radionuclide release from the reactor containment to the environment during accidents initiated while the reactor is at power and the containment is initially intact follow three paths:

- leakage from a pressurised containment;
- flow through an interfacing system such as a ruptured steam generator tube; and
- catastrophic release from a ruptured containment.

The minimum release of radioactivity to the environment during a severe reactor accident will be that which occurs by leakage from a containment that is otherwise intact. Reactor containments are typically robust structures that are tested routinely for leakage at pressures up to and even exceeding the design pressure. Allowable leakages are usually mandated by regulation to be at most a few percent per day. Leakages found in tests are often less than 0.5% per day [C-1]. What leakage is found is usually attributed to the numerous penetrations of the containment for pipes, electrical conduits, and personnel access. It is, however, common to find that extensive preparation for the leakage test was necessary. Without these preparations, unacceptably large leak rates would be encountered because of errors in sealing the containment and errors in the maintenance or the modifications of penetrations.

Indeed, the operational record of nuclear power plant operation has shown several instances of failure of the containment isolation function [C-2]. The probability that the containment leakage rate exceeds the allowable limit at any given time during plant life was assessed and shown to be relatively high. In most of the cases the leakage rate was not much higher than the allowable limit, but in some cases it was such that the potential environmental consequences of an accident would have been large. Therefore, leak rates in excess of the design base during severe accidents cannot be discounted and accident management strategies to mitigate these leaks have to be implemented.

The first priority is obviously to detect and plug the leaks as soon as possible. Different detection methods, capable to monitor the leaktightness of the containment during reactor operation, are possible [C-2]. For example, in Belgium and in France, the containment integrity of the PWRs with a large dry containment is routinely monitored. In normal operation, the pressure in the reactor building has a tendency to increase owing to the leakage of the compressed air system. If one measures the flow of incoming air, the pressure, the temperature and the humidity in the building, it is possible to calculate the leakage rate. Tests have shown that a hole of 0.7 cm in diameter can be detected in one or two days.

A complementary measure to reduce the consequences of containment isolation failures (either due to preexisting openings or to a failure of the containment isolation system) is to prepare an accident procedure to help in the detection and the isolation of an opening, after the accident has taken place. In France for example, the U2 procedure addresses the search for and processing of abnormal containment leaktightness defects after an accident has caused a fuel degradation and/or a primary system defect [C-2]. The generic severe accident management guidelines (SAMG) developed by various owners' groups also address the issue.

Bounding analyses of the radionuclide release by leakage from intact containments usually assume that the radionuclides behave like gases and follow without attenuation flow paths through the containment structure. In reality, of course, many of the radionuclides of most concern will be suspended in the containment atmosphere as aerosol particles. These particles need not pass through leak paths unattenuated. They can be trapped by diffusion, interception or impaction on surfaces in the leak path [C-3]. It is often observed that deposition of particles along a gas flow path distorts the flow sufficiently to facilitate further deposition of particles. There is, then, a tendency for leak paths to plug with aerosols and various correlations of the time required to plug leak paths have been formulated. Accident management efforts could be considered additional measures of defense in depth against the accidental release of radioactive materials.

Flow of radioactive gases and aerosol through interfacing systems are hallmarks of the most risk significant reactor accidents. Such modes of release do merit attention in the development of accident management strategies. The most direct way to attenuate release of radioactivity by this means is to close the pathway created by the rupture of an interfacing system. In the case of a steam generator tube rupture accident, this involves closing safety relief valves on the secondary side of the steam generator. This direct measure is not always possible because of mechanical features of the system or because of the intense radiation fields that are created along the flow path. Most alternative approaches have focused on reducing the reactor coolant system pressure to reduce the driving force for release of radionuclides and submerging the point of release under water. For example, efforts could be made to flood the secondary side of a steam generator containing a ruptured tube. Certainly, flooding the secondary side is the strategy for release mitigation adopted for the Temelín VVER-1000. At Sizewell, an engineered system to redirect flows from the secondary side of the steam generator to a point below the surface of the spent fuel pool has been considered again to achieve submergence of the point of release. Submergence provides attenuation entirely akin to the attenuation achieved by sparging gases through water pools (see Chapter V). That is, aerosol particles are removed by a combination of diffusion, inertial impaction and sedimentation processes. Keeping the water pool basic (high pH) would also aid the removal of gaseous forms of iodine. A more ambitious strategy would be to engineer systems that would permit diversion of flows to other bodies of water such as the spent fuel storage pools.

When flooding is not possible, it can be imagined that makeshift filters could be packed around the points of radionuclide release. Certainly, this was part of the strategy in mind when sand and clay were dumped into the Chernobyl core during the accident in 1986 [C-5]. Post accident assessments of the accident management effort at Chernobyl, which was largely unsuccessful, even suggested that filtration materials might be stockpiled as an accident management measure in anticipation of future accidents.

Containment failure may be the eventual outcome of an unmitigated reactor accident. Failure need not be catastrophic. In fact, general cracking is expected when concrete containments are overpressurised. There may be some natural mitigation of radionuclide release following containment rupture. Aerosols may deposit along the flow pathway. Steam condensation upon depressurisation may create droplets that will sweep radioactive aerosol particles out of the atmosphere [C-6]. A persistent source term concern associated with containment rupture is that water pools within the containment will undergo "flash" generation of steam. The flow of steam might then be sufficient to entrain radionuclide contaminated water droplets that are carried out of the containment. Repeated experimental and analytical investigations of this possibility have shown that significant radionuclide releases by this mechanism are possible only for catastrophic containment rupture with very large depressurisation rates [C-7]. For more probable situations involving slow depressurisation of containment, the releasable inventory of radionuclides that is most important is that suspended in the containment atmosphere and the iodine that slowly partitions from water pools.

Plants with a double containment are offered some additional ways to cope with containment releases. With an intact containment, the space between both containments may be held subatmospheric and the leakages collected and filtered before release. The ventilation system used for that purpose usually has a large design margin and can therefore process a primary containment leakage rate far in excess of the normal value (if AC power is available).

It is only when the primary containment completely fails, that the secondary containment may be pressurised. Secondary containments are usually strong structures, designed to cope with external events, but their pressure retaining capability is in most cases not verified. It may therefore be expected that large leaks will develop to the outside if it comes under pressure. Even so, the longer leakage path will ensure some additional retention compared with a plant with single containment.

Some plants, equipped with a steel primary containment, are also contemplating or have installed a spray system in the space between the two containments (e.g. at Loviisa [C-8]). The main purpose of such a system is to cool the primary containment in case all other means are lost, but of course it will have a scrubbing effect on the accidental releases.

The main effect of the external spray system on the source term is, of course, maintaining the containment integrity and thus avoiding major leakages into the environment. The scrubbing of the aerosols from the space outside the steel shell (called outer annulus in Loviisa) is thought to be minor as the external spray sweeps only a fraction of the outer annulus gas space. Scrubbing would be efficient in those sequences only where the release route goes through the topmost part of the outer annulus. Another mechanism affecting the source term with the external spray is the enhanced steam condensation in the containment dome which results in more efficient aerosol removal from the containment atmosphere by diffusiophoresis than without the external spray. However, these mechanisms can reduce the source term significantly only in sequences with intact containment, in which the source term is already very low.

Accident management measures to mitigate release following containment rupture will be difficult to design simply because the rupture location will be difficult to predict. For this reason, concepts such as filtered vented containment designs have been devised. These designs enforce a relief direction for radionuclide-laden gases before containment failure can occur [C-9]. Perhaps, the most famous of the filter systems is the large gravel tunnel filter at the Barsebäck station [C-10]. Many other technologies are available for the filtration of aerosol-laden gases including:

- deep metal fiber filters;
- gravel bed filters;
- washed Venturi scrubbers; and
- electrostatic precipitators.

Typical mechanisms for aerosol removal from gas streams by filters are diffusion to surfaces, interception and impaction. Very large particles can be removed by gravitational settling. These mechanisms are quite dependent on the particle size and it is usually found that conventional filters have a minimum in filter efficiency for particles in a narrow size range less than 1 µm. When the gas is hot relative to the filter, thermophoresis can enhance particle removal. When the aerosol laden gas stream contains elevated concentrations of steam that condenses within the filter, diffusiophoresis will enhance particle removal. These phoretic enhancements of filtration are attractive because filtration efficiencies by these mechanisms are not especially dependent on the aerosol particle size. Washed Venturi scrubbers involve the injection of water droplets into the aerosol laden gas and these water droplets act much like spray water droplets to remove aerosol particles. Electrostatic precipitation is, in principle, a very attractive decontamination process, but it is difficult to assure that the necessary power will be available to operate the precipitators under accident conditions.

The most important constraints on the design of filtered venting systems are the mass loads and the thermal loads on the system. Mass loads are difficult to predict because of the large amount of nonradioactive aerosol material that can be created in a severe reactor accident. Thermal loads arise from the accumulation of radionuclides in the filter and can create temperatures sufficient to revaporise volatile species of iodine and cesium from the filter. Two strategies have emerged to treat these constraints. One strategy is to make the reactor containment large and strong so that filtered venting is not needed until very late in the accident when most of the aerosol mass has been removed from the reactor containment atmosphere by natural processes or engineered systems [C-11]. The other strategy is to make the filter

system large. Wetted systems are especially attractive since water washing of the filter medium prevents plugging as well as removing heat generated by the radionuclides [C-12]. Wet surfaces also prevent particle bounce as a mechanism for particle penetration of the filter. Wetted systems do require additional features to remove radionuclide contaminated water mist entrained in the flow through the filter.

To illustrate the concepts developed in this chapter, the severe accident management guidelines, aimed specifically at reducing the containment releases to the environment, as implemented at the Tihange unit 2 nuclear power plant, are taken as an example. Tihange 2 is a PWR with a Framatome NSSS and a large dry double concrete containment designed by Tractebel. The first step of the strategy to reduce environmental releases is to identify possible leakage paths by means of the radioactivity surveillance instrumentation.

If the leak is trough a containment penetration, the penetration should be isolated (e.g. by closing a valve in the leakage path). If this is not sufficient or not possible, the containment spray system should be started (if not yet done) in order to (1) reduce the airborne fission products inventory in the containment, and (2) reduce the driving force for the leak.

If the leak is through a steam generator, several measures are possible:

- the steam generator could be filled with water in order to scrub the radioactive products;
- the steam generator could be isolated (but in that case the steam pressure has to remain below the safety valves set point);
- the steam could be dumped to the condenser (if available). Fission products would be trapped in the condenser. Even though this measure contaminates equipment located in the machine hall, this may be preferable to an uncontrolled release to the outside.

If the leak ends in the auxiliary building, again several measures are possible:

- isolate the system responsible for the leak; this may not be possible if the system is essential for managing the accident (e.g. safety injection or containment spray);
- start the ventilation system (if not yet done) in order to filter the leak and discharge it through the stack, thereby reducing its radiological impact;
- start a spray system in the rooms along the leakage path (e.g. the fire fighting sprinklers). This strategy is not used at Tihange 2 because it requires system modifications that were not implemented.

VII.2 Mitigation of release during shutdown accidents

Accident management measures need to be considered for accidents initiated while the nuclear plant is in shutdown operations because many of the normal safety systems and barriers to radionuclide release are not available. Releases through auxiliary buildings at rather slow rates do provide opportunities for source term attenuation using sprays, water pools and filters. In light of the high risks that can be posed by accidents under shutdown conditions, it is surprising that so little attention has been devoted to the radionuclide source terms that can come from such accidents. There has been little attention given to attenuating these source terms by accident management measures.

VII.3 Annotated Bibliography on Mitigation of Release from Containment

C-1. F. Robledo, Containment Bypass and Leaktightness, NEA/CSNI/R(95)25, Committee on the Safety of Nuclear Installations, OECD Nuclear Energy Agency, France, October 1995.

The paper presents a discussion of leakage in several containments and the local leakage rate testing.

C-2. B. De Boeck, **Inadequate Isolation of Containment Openings and Penetrations**, CSNI Report 179, Committee on the Safety of Nuclear Installations, Paris, 1990.

This report reviews international experience regarding the assessment of the probability of preexisting openings in the containment, and describes methods to detect them, either during reactor operation or after an accident has taken place.

C-3. C. Coroli, N. Coulon, D.A.V. Morton, and M.M.R. Williams, "Theoretical and Experimental Investigations of the Leakage of Steam, Gas, and Aerosols Through Narrow Cracks and Capillaries", **Reinforced concerted action on reactor safety - FISA 95 - EU research on severe accidents,** EUR 16896 en, G. Van Goethem, W. Balz, and E. Della Loggia, editors, Directorate-General for Science, Research, and Development, European Communities, Brussels, Belgium, 1996.

This paper presents a discussion of theoretical and experimental investigations of the efficiency of aerosol transport through narrow structures such as gaps around containment penetrations or cracks in containment caused by over pressurisation.

C-4. J. Li, D. Leaver, and J. Metcalf, "Aerosol Retention During An Unisolated Steam Generator Tube Rupture Severe Accident Event", **Proceedings on the Fifth International Topical Meeting On Nuclear Thermal Hydraulics, Operations and Safety**, Beijing, China, April 13-16, 1997.

This paper argues that the natural attenuation of aerosol in flows through the secondary sides of steam generators is large and that risks associated with steam generator tube rupture accidents may be smaller than has been estimated in past risk analyses.

C-5. M. W. Jankowski, D. A. Powers, and T. S. Kress, "Onsite Response to the Accident at Chernobyl (Accident Management)", **Nuclear Safety 28**, (1987) 36.

The paper describes the heroic efforts undertaken in the Soviet Union to mitigate the releases of radionuclides during the Chernobyl accident.

C-6. C.D. Leigh, *et al.*, **Analyses of Plume Formation, Aerosol Agglomeration and Rainout Following Containment Failure**, NUREG/CR-4222, SAND84-2581, Sandia National Laboratories, Albuquerque, NM, August 1986

Analyses are presented of possible natural processes to mitigate the release of radionuclides following containment rupture.

- C-7a. R. Borkowski, H. Bunz, and W. Schöck, **Resuspension of Fission Products During Severe Accidents in Light Water Reactors**, XII/978/84-EN, Kernforschungszentrum Karlsruhe, Karlsruhe, Germany, November 23, 1984.
- C-7b. J.E. Brockmann, "Range of Possible Resuspension and Reentrainment of Fission
- Products during Containment Depressurization", Appendix G, R.J. Lipinski et al., **Uncertainty in Radionuclide Release Under Specific LWR Accident Conditions-Volume II TMLB' Analyses**, SAND84-0410, Volume 2, Sandia National Laboratories, Albuquerque, NM, February 1985.
- C-7c. Industry Degraded Core Rulemaking, **Resuspension of Deposited Aerosols Following Primary System or Containment Failure**, Atomic Industrial Forum, Bethesda, MD, 1984.
- C-7d. T. Kudo, N. Yamano, K. Moriyama, Y. Maruyama, and J. Sugimoto, "Experimental Study of Aerosol Reentrainment from Flashing Pools in ALPHA Program", Proceedings of the 3rd International Conference on Containment Design and Operation, Toronto, Canada October, 1994.

These reports of experiments and analyses show that resuspension of fission products other than iodine from water pools during containment depressurisation is important only when there is very rapid depressurisation of the containment such as might occur when the rupture diameter exceeds about 10 m.

C-8. H. Tuomisto, *et al.*, "External spray cooling of the Loviisa containment", **Proceedings of the Specialist Meeting on Selected Containment Severe Accident Management Strategies**, CSNI Report NEA/CSNI/R(95)3, Committee on the Safety of Nuclear Installations, Paris, 1995.

The paper describes the external spray containment cooling system installed at the Loviisa nuclear power plant, the reasons why the system was installed, and the design calculations and supporting experiments performed to validate the concept.

C-9. **Proceedings of a Specialists' Meeting on Filtered Containment Venting Systems**, CSNI Report 148, Committee on the Safety of Nuclear Installations, Paris, 1988.

Proceedings of this conference provide a good indication of the state of engineering technology for the development of filtered venting systems.

C-10. A. Hedgran, et al., FILTRA Final Report, November, 1982.

This report summarizes the work done to obtain basic performance data for a stone-filter and the design of the FILTRA system for the two reactors at Barsebäck.

C-11. H.-G. Dillmann, H. Pasler, and J.G. Wilhelm, "Filtered Venting for German Power Reactors", **Nuclear Technology**, **92**(1990)40.

The paper presents a detailed discussion of deep-bed stainless steel fiber filters for reactors.

German

C-12. L. Lindau and K. Ellisson, "Filtered Containment Venting in Sweden", **Proceedings of the Twentieth DOE/NRC Nuclear Air Cleaning Conference**, NUREG/CP-0098, pp.695-707, U.S. Nuclear Regulatory Commission, Washington, D.C., 1988.

The paper describes the multi-Venturi scrubber system developed in Sweden for filtered venting of reactor containments. Additional details can be found in E. Söderman, editor, **Severe Accident Analysis in Sweden - Methods and Results, RAMA III Final Report**, RAMA-III 89-02, ISBN 91-7010-116-7, December 1989.

APPENDIX A

Information provided by

Belgium
Canada
Czech Republic
Finland
France
Germany
Japan
Republic of Korea
Spain
The Netherlands
Sweden
Switzerland
USA

regarding specific features of currently operating nuclear power plants

Korea:

Plant Name	Туре	Designer	Operational date	Thermal power (MWt)	Containment Type ²⁾	Free volume (m³)	Sprays ³⁾	Containment Sump chemical Additives ⁴⁾	Filters ^{5),6)}	Containment Venting filters
Kori-1	PWR	W	1978. 4. 29	1,723.5	Large Dry (Dual)	41,100	750 m ³ /hr Additive: 30% NaOH	-	HEPA: containment recirculation, 52,670 m ³ /hr (99.99%)	-
Kori-2	PWR	W	1983. 7. 25	1,876	Large Dry (Dual)	40,800	538 m ³ /hr Additive: 30% NaOH	-	-	Annulus negative pressure control sys.: 5,100 m³/hr (HEPA/Charcoal: 99.97/99.90%)
Kori-3 & 4	PWR	W	1985. 9. 30/ 1986. 4. 29	2,775	Large Dry	60,900	1,363/1,658 m³/hr (injection/recirculation) Additive: 27~30% NaOH	-	HEPA/Carbon: Fuel Bldg. Emergency Exhaust, 8,495 m³/hr (99.97/95%)	-
Yonggwang- 1&2	PWR	W	1986. 8. 25 1987. 6. 10	2,775	Large Dry	60,900	Same as Kori-3 & 4	-	Same as Kori-3 & 4	-
Yonggwang- 3&4	PWR	ABB-CE	1995. 3. 31 1996. 1. 1	2,815	Large Dry	77,220	1,590 m ³ /hr Additive: 30~35% N ₂ H ₄	TSP: 24,970 kg pH: 7.0~8.5	HEPA : ECCS equip. Room Exhaust, 10,200 m³/hr	-
Ulchin-1&2	PWR	Framatome	1988. 9. 10 1989. 9. 30	2,665/ 2,775	Large Dry	49,400	850~970 m³/hr Additive: 25~32% NaOH	-	Absolute/Charcoal: Peripheral rooms & Fuel Bldg. Ventilation 3,600m³/hr (99.95/99%)	-
Ulchin-3 (&4)	PWR	KOPEC	1998. 8. 11	2,815	Large Dry	77,220	2,271 m³/hr No additive	TSP:24,970 kg pH: 7.0~8.5	Same as Yonggwang-3&4	-
Wolsong-1	PHWR	AECL	1983. 4. 22	2,064	Single bldg. with dousing water storage tank	48,478	Dousing water: 2,050m ³ , 24,494 t/hr Additive: N ₂ H ₄ 50~100 mg/kg H ₂ 0	-	-	Carbon bed: filtered discharge, 16,920 m³/hr (99.5%)
Wolsong-2, 3 (& 4)	PHWR	AECL	1998. 7. 1	2,061.4	Single bldg. with dousing water storage tank	48,000	Dousing water: 1,559 m³, 24,494 t/hr Additive: N ₂ H ₄ 50~100 mg/kg H ₂ 0	TSP: 100 kg pH: 9.0~10	-	-

Plant Name	Passive iodine removal	lodine mitigation measures implemented/ planned	Measurement of airborne iodine (gaseous and particulate form) activity in containment	Measurement of controlled iodine (gaseous and particulate form) release into environment	Other AM measures to mitigate airborne iodine (in gaseous/particulate form) activity in containment	Comments
Kori-1	Plate-out	Spray additive	Post-accident air sampling and on-line radiation monitoring	on-line radiation monitoring	-	
Kori-2	Plate-out	Spray additive	Same as above	Same as above	-	
Kori-3 & 4	Plate-out	Spray additive	Same as above	Same as above	-	
Yonggwang- 1&2	Plate-out	Spray additive	Same as above	Same as above	-	
Yonggwang- 3&4	Plate-out	TSP in Sump, Spray additive	Same as above	Same as above	-	
Ulchin-1&2	Plate-out	Spray additive	Same as above	Same as above	-	
Ulchin-3 (&4)	Plate-out	TSP in Sump, Spray without additive	Same as above	Same as above	-	
Wolsong-1	Plate-out	Spray additive	Same as above	Same as above	-	
Wolsong-2, 3 (&4)	Plate-out	TSP in Sump, Spray additive	Same as above	Same as above	-	

¹⁾ The plants in parenthesis are under construction. In addition, Yonggwang units 5&6, which are similar to the Ulchin-3 (&4), are also being constructed.

Dual containment consists of a steel shell and a shield building between which annulus exists.

The spray flow rates are the sum of those of all trains.

⁴⁾ TSP: Tri-sodium phosphate

The above characteristics are only for post-accident condition. As for normal operation, all plants have HEPA and charcoal filters. Every plant has control room emergency makeup system for the protection against LOCA, which consists of HEPA filter and charcoal adsorbers.

⁶⁾ Filters for the systems outside the containment can also provide partial contributions to the removal of iodine following LOCA.

Finland:

Plant Name	Туре	Designer	Operatio nal date	Thermal power (MWt)	Containment type	Free volume (m³)	Sprays ¹⁾	Containment Sump chemical Additives	Filters	Containment Venting filters
Loviisa 1 & 2 (Lo1 & Lo2)	VVER- 440/213	V/O Atomenergo export (Russia)		1500	Westinghous e ice condenser containment	Gas volume : 57 000 m³ Maximum sump volume: 2000 m³	Lo1: 566 kg/s Lo2: 497 kg/s N ₂ H ₄ : max. 106 mg/kg (with the spray flow rate of 500 kg/s) KOH: max. 423 mg/kg (with the spray flow rate of 500 kg/s) max. 423 mg/kg)	Maximum sump water volume: N_2H_4 : 0.77120 mg/kg (injected to spray water) KOH: 0.24500 mg/kg (injected to spray water) $Na_2O(B_2O_3)_2$:3.8 g/kg (from ice) $B(OH)_3$: 6.27.1 g/kg pH: 89 (expected in the containment) pH:≤ 6 (in the bypass sequences when ice and other additives not involved) Minimum sump water volume (primary circuit+accumulators+ice): N_2H_4 : 1.4 mg/kg (from the primary coolant; decomposed into NH ₃) KOH: 0.43.4 mg/kg (from the primary coolant) $Na_2O(B_2O_3)_2$:6.7 g/kg (from ice) $B(OH)_3$: 1.63.4 g/kg pH: 89 expected in the containment) Bypass sequences, when ice and other additives not involved: pH: ≤ 6		None (containment venting not considered as a SAM measure for the Loviisa NPP).

¹⁾ The actual concentrations depend on the operation of the spray water and chemical additive solution pumps. The concentrations represent the situation before recirculation phase. During recirculation chemicals can still be fed into the spray system, giving increase in the concentrations.

From the so called outer annulus (the space outside the containment between the containment steel shell and the reactor building concrete wall) air is sucked into the air ventilation stack. In case of the leakage into the outer annulus the air flow is switched to pass through the filter line with four sequential filters: 1st prefilter, 2nd HEPA filter, 3rd KI impregnated active carbon filter, 4th HEPA filter. Absolute capacities of the filtering systems are not known.

The minimum requirements for the efficiencies of new HEPA filters is 99.97%. The efficiency is expected to increase with older HEPA filters.

The minimum requirements for the efficiencies of active carbon filters in use is 99% for removal of CH₃I. The efficiency of molecular iodine would be thus at least 99.9%.

The incoming air to the main control room and the plant emergency management room is also filtered with similar requirements than presented above. The systems include prefilters, HEPA filters and active carbon filters. In the system for the main control room the HEPA and active carbon filters are in the same box.

²⁾ Only the systems designed to be involved with release mitigation during a reactor accident are presented here, and thus the filtering for the containment under pressure and ventilation systems are not considered.

Plant Name	Passive iodine removal	lodine mitigation measures implemented/ planned	Measurement of airborne iodine (gaseous and particulate form) activity in containment	Measurement of controlled iodine (gaseous and particulate form) release into environment	Other AM measures to mitigate airborne iodine (in gaseous/ particulate form) activity in containment (3)	Comments
Loviisa 1 & 2 (Lo1 & Lo2)	Borax in the ice condensers makes the ice rather basic, and thus the ice removes quite efficiently also gaseous iodine (I ₂) from the gas flows.	Borax in the ice condenser ice (implemented). Injection of N ₂ H ₄ and KOH into the spray water (implemented).	Activity concentrations of different iodine isotopes in the containment gas phase can be measured from the samples taken from the containment air. No chemical speciation is measured.	The filter line in the air ventilation stack is equipped with sampling cartridges. The collecting efficiency of CH ₃ I is known for the sampling, and the efficiency for other iodine species can be estimated. From these samples the release of different iodine species into the environment can be evaluated.		

³⁾ In order to mix the hydrogen produced in zirconium oxidation throughout the containment, ice condenser doors are forced open (planned). This enables a global convective loop flow between the steam generator room and the upper compartment through the ice condensers. The loop flow carries along both particulate and gaseous iodine, which are partially removed from the gas flowing through the ice condenser sections. Also, due to high relative humidity in the steam generator room and hygroscopicity of the particulate iodine species, iodine in aerosol form is expected to deposit in the steam generator room more efficiently than in the upper compartment of the containment

Spain:

Plant Name	Туре	Designer	Operational date	Thermal power (MWt)	Containment type	Free volume (m³)	Sprays	Containment Sump chemical Additives	Filters	Containment Venting filters
José Cabrera	PWR	W	1968	510	Dry	23393	no internal but external spay system		Containment Atmosphere Emergency Filtration System (CAEFS): - One fan with a flow capability of 1250 m³/min - One unit for fission products removal from containment: - Electrical heater Humidity separator Prefilters HEPA filters: (efficiency against DOP: 99,97%) Active Carbon Filters: 3 cartridges of 50, 8 mm thick each., Efficiency: 95-99%	none
Santa María de Garoña	BWR	GE	1971	1381	Mark I	6153 (1)	drywell and torus spray system, no additives	no additives in suppression pool	Active Carbon Filters: Efficiency: 95-99%	none
Almaraz 1,2	PWR	W	1980/1983	2696	Dry	59465	- two trains: Each train with flow capability of 3120 gpm in the injection phase and 3500 gpm in the recirculation phase Spray Additive System: - one tank containing 4850 gallons of solution of NaOH with concentration of 30 wt %.		Active Carbon Filters: Efficiency: 95-99%	none

Plant Name	Туре	Designer	Operational date	Thermal power (MWt)	Containment type	Free volume (m³)	Sprays	Containment Sump chemical Additives	Filters	Containment Venting filters
Cofrentes	BWR	GE	1985	2894	Mark III	35345 (2)	spray system, no additives	no additives in the suppression pool	Active Carbon Filters: Efficiency: 95-99%	none
Ascó 1,2	PWR	W	1983/1985	2696	Dry	62092	2 trains, each one with one pump with flow capability: 1500 gpm	baskets with trisodium phosphate (Na ₃ PO ₄) in containment sump maintaining the containment sump pH at 7.2	Active Carbon Filters: Efficiency: 95-99%	none
Trillo	PWR	KWU-Siemens	1988	3010	Dry	56811	no spray	no additives	Active Carbon Filters: Efficiency: 95-99%	none
Vandellós II	PWR	W	1988	2785	Dry	62578	- two trains: Each train with flow capability of 3120 gpm in the injection phase and 3500 gpm in the recirculation phase Spray Additive System: - one tank containing 4850 gallons of solution of NaOH with concentration of 30 weight %.		Active Carbon Filters: Efficiency: 95-99%	none

⁽¹⁾ Drywell 3293 m³; torus 2858 m³.

⁽²⁾ Drywell 6822 m³; wetwell 28253 m³

Plant Name	Passive iodine removal 3)	lodine mitigation measures implemented/ planned	Measurement of airborne iodine (gaseous and particulate form) activity in containment	Measurement of controlled iodine (gaseous and particulate form) release into environment	Other AM measures to mitigate airborne iodine (in gaseous/ particulate form) activity in containment	Comments
José Cabrera	none	none	iodine activity measured	iodine activity measured	none	
Santa María de Garoña	none	none	iodine activity measured	iodine activity measured	none	
Almaraz 1,2		none	iodine activity measured	iodine activity measured	none	
Cofrentes	none	none	iodine activity measured	iodine activity measured	none	
Ascó 1,2	none	none	iodine activity measured	iodine activity measured	none	
Trillo	none	none	iodine activity measured	iodine activity measured	none	
Vandellós II	none	none	iodine activity measured	iodine activity measured	none	

France:

Plant Name	Type	Designer	Operational date	Thermal	Containment	Free	Sprays	Containment	Filters	Containment 1)
CP0 series (6 units) 900 MWe	PWR	FRAMATOME	Ranging from March 1977 to July 1979	power (MWt) 2'785 (except FSH: 2 660)	type Large dry Single wall with steel liner	volume (m³) 47'000	Two files	sump chem. additives Boric acid and soda (high pH)	(Ventilation systems) Two files of HEPA filters upstream of the stack	venting filters Internal metallic pre-filter + extern sand-bed filter
CP1 series (18 units) 900 MWe	PWR	FRAMATOME	Ranging from February 1980 'to May 1983	2785	Large dry Single wall with steel liner	51'000	Two files	Boric acid and soda (high pH)	Two files of HEPA filters upstream of the stack	Internal metallic pre-filter + extern. sand-bed filter
CP2 series (10 units) 900 MWe	PWR	FRAMATOME	Ranging from January 1981 to October 1984	2785	Large dry Single wall with steel liner	51'000	Two files	Boric acid and soda (high pH)	Two files of HEPA filters upstream of the stack	Internal metallic pre-filter + extern sand-bed filter
P4 series (8 units)	PWR	FRAMATOME	Ranging from	3'817	Large dry	81'000	Two files	Boric acid	Two files of	Internal metallic
1300 MWe			May 1984 to June 1986		Double wall w/o liner			and soda (high pH)	HEPA filters upstream of the stack	pre-filter + extern. sand-bed filter
P'4 series (12 units) 1300 MWe	PWR	FRAMATOME	Ranging from October 1986 to November 1992	3'817	Large dry Double wall w/o liner	70'000	Two files	Boric acid and soda (high pH)	Two files of HEPA filters upstream of the stack	Internal metallic pre-filter + extern. sand-bed filter
N4 series (3 units, plus one to be started) 1450 MWe	PWR	FRAMATOME	Ranging from 1996 to 1999	4'250	Large dry Double wall w/o liner	73'000	Two files	Boric acid and soda (high pH)	Two files of HEPA filters upstream of stack	Internal metallic pre-filter + extern. sand-bed filter

¹⁾ Internal metallic pre-filter retained efficiencies: factor 10 for aerosols (before filter plugging); factor 1 for l₂, ICH₃ and noble gases

^{2) (}Upstream liaison piping + Sand-bed filter) retained efficiencies: factor 100 for aerosols, factor 10 for I₂, factor 1 for ICH₃ and noble gases

Plant Name	Passive iodine removal	lodine mitigation measures	Measurement of airborne iodine activity in containment	Measurement of controlled iodine release into environment	Other AM measures to mitigate iodine activity in containment
CP0 series (6 units)900 MWe	Natural Processes only	Alcaline borated spray water	Global dose rate measurement in the containment	Between the sand-bed filter and the special duct inside the stack (*)	None
CP1 series (18 units) 900 MWe	Natural Processes only	Alcaline borated spray water	Global dose rate measurement in the containment	Global dose rate Between the measurement sand-bed filter	
CP2 series (10 units) 900 MWe	Natural Processes only	Alkaline borated spray water	Global dose rate measurement in the containment	Between the sand-bed filter and the special duct inside the stack (*)	None
P4 series (8 units) 1300 MWe	Natural Processes only	Alcaline borated spray water	Global dose rate measurement in the containment	Between the sand-bed filter and the special duct inside the stack (*)	None
P'4 series (12 units) 1300 MWe	Natural Processes only	Alcaline borated spray water	Global dose rate measurement in the containment	Between the sand-bed filter and the special duct inside the stack (*)	None
N4 series (3 units, plus one to be started) 1450 MWe	Natural Processes only	Alcaline borated spray water	Global dose rate measurement in the containment	Between the sand-bed filter and the special duct inside the stack (*)	None

Belgium:

Plant name	Type: PWR	Designer	Op. date	Thermal power (MWth) (core/total)	Containm	ent type	Free volume (min/max)	Tihange 1: 2700ppm)		Qualified containment cooling (Doel)	Containment sump chemical additives
	nb loops				inner containment	outer containment	(m3)	Capacity (m3/h)	additive (NaOH) 20-30 %	Capacity (m3/h)	NaOH
Doel 1 and Doel 2, twin units	2	W	1974	1187/1192	metallic sphere	reinforced concrete	42000 / 43000	injection phase only: 2 x 120 (2 x 50%) (+ 2 shared backup units)	no	recirculation phase only 4 X 80000	no
Doel 3	3	Framatome	1982	3054/3064	prestressed concrete with metallic liner	reinforced concrete	60113 / 64000	3 x 530 (3 x 50%)	recirculation phase	6 x 125000	recirculation phase pH (7;9.5)
Doel 4	3	W	1985	2988/3000	prestressed concrete with metallic liner	reinforced concrete	59422 / 60964	3 x 530 (3 x 50%)	recirculation phase	6 x 125000	recirculation phase pH (7,9.5)
Tihange 1	3	Framatome	1975	2867/2873	prestressed concrete with metallic liner	reinforced concrete	68250 / 69850	injection phase: 2 x 400 recirculation phase: 2 x 500 [+ 2 safeguard pumps (inj. & recirc.): 500] (2 x 50% + 2 X 50%)	recirculation phase	-	recirculation phase pH (8.5;10.5)
Tihange 2	3	Framatome	1982	2895/2905	prestressed concrete with metallic liner	reinforced concrete	67200 / 69100	3 x 520 (3 x 50%)	recirculation phase	-	recirculation phase pH (8,10)
Tihange 3	3	W	1985	2988/3000	prestressed concrete with metallic liner	reinforced concrete	71812 / 72900	3 x 555 (3 x 50%)	recirculation phase	-	recirculation phase pH (8,10)

Plant name	Filters (PF=prefilter, AF=absolute filter D = demister)	er, ACF=active carbon filter,	Containment venting filters	Passive iodine removal	lodine mitigation measures implemented/ planned	Measurement of airborne iodine (gaseous and particulate form) activity		
	depressurization system of the annulus	ulus (recirculation)			containment + annulus	stack		
Doel 1 and Doel 2, twin units	2 x 85 m3/h (2 x 100%) AF+ACF AF 2500 m3/h, 99% (> 0.3µ), ACF 1400m3/h (1%KI) I: 90%	4 x 5000 m3/h PF+AF+ACF (4 x 50%) PF 2500 m3/h, 95% (> 6 μ), AF 2500 m3/h, 99% (> 0.3μ), ACF 1700m3/h (1%KI) I 90%	-	-	-	gamma spectrometry of samples + continuous spectrometry	gamma spectrometry of samples + continuous spectrometry	
Doel 3	2 x 300 m3/h (2 x 100%) PF+AF+ACF+AF PF 85%, AF 99% ACF I: 90%	3 x 13000 m3/h (3 x 50%) PF+AF+ACF PF 85%, AF 99% ACF I: 90%	-	-	-	gamma spectrometry of samples + extended scale measuring channel (NaI)		
Doel 4	2 x 300 m3/h (2 x 100%) PF+AF+ACF+AF PF 85%, AF 99% ACF I 90%	3 x 13000 m3/h (3 x 50%) - PF+AF+ACF PF 85%, AF 99% ACF I 90%	-	-	-	gamma spectrometry of samples + extended scale measuring channel (NaI)		
Tihange 1	2 x 1700 m3/h D+PF+ACF+AF ICH3: 90% I2: 95%	2 x 5100 m3/h D+PF+FA+ACF ICH3: 90% I2: 95%	-	-	-	gamma spectrometry of samples	gamma spectrometry of samples	
Tihange 2	3 x 2150 m3/h (3 x 50%) -ACF+AF AF 99% ACF (I2,ICH3) 94%	3 x 9000 m3/h (3 x 50%) D+PF+AF+ACF AF 99% ACF (I2,ICH3) 94%	-	-	-	gamma spectrometry of samples	gamma spectrometry of samples	
Tihange 3	3 x 2150 m3/h (3 x 50%) - ACF+AF AF 99% ACF (I2,ICH3) 94%	3 x 9500 m3/h (3 x 50%) D+PF+AF+ACF AF 99% ACF (I2,ICH3) 94%	-	-	-	gamma spectrometry of samples	gamma spectrometry of samples	

No containment venting filters, no passive iodine removal, no controlled iodine release into environment, no other AM measures to mitigate airborne iodine activity in containment

Sweden:

Plant Name	Туре	Designer	Operational date	Thermal power (MWt)	Containment type	Free volume (m³) (1,2)	Sprays ⁽³⁾ (kg/s)	Containment Sump chemical Additives (4)	Filters ⁽⁵⁾	Containment Venting filters ⁽⁶⁾
Ringhals 1	BWR	ABB	1976	2500	PS	4976 2989	300			multiventuri system, DF: 100
Ringhals 2	PWR	WESTINGHOUSE	1975	2660	Large dry	51000	500			multiventuri system, DF: 500
Ringhals 3	PWR	WESTINGHOUSE	1981	2783	Large dry	51000	500			multiventuri system, DF: 500
Ringhals 4	PWR	WESTINGHOUSE	1983	2783	Large dry	51000	500			multiventuri system, DF: 500
Forsmark 1	BWR	ABB	1981	2928	PS	4315 2555	400			multiventuri system, DF: 100
Forsmark 2	BWR	ABB	1981	2928	PS	4315 2555	400			multiventuri system, DF: 100
Forsmark 3	BWR	ABB	1985	3300	PS	5857 2850	400			multiventuri system, DF: 100

- 1) Containment (gas volume)
- 2) Drywell, wetwell (gas/pool volume), for BWR: total free gas volume/wetwell gas volume
- 3) Flow capacity (injection phase/recirculation phase), chemical additive (if yes, what, concentration): BWRs contain LiOH to maintain pH at 8-8.5, this will act as a chemical additive to the sump water. PWRs: trisodiumphosphate is added during the recirculation phase to increase pH and increase absorption of iodine
- 4) Chemical name, expected concentration, expected pH: BWRs contain LiOH to maintain pH at 8-8.5, this will act as a chemical additive to the sump water
- 5) Type (e.g., Prefilters, HEPA filters, active carbon filters), designed adsorbing capacity and efficiency: BWRs: a delay system exists after turbine condenser equipped with active coal filters. Also HEPA filters are used in BWRs
- 6) The scrubbers are designed with the given Decontamination Factors (DF), The real DFs are still higher

Plant Name	Passive iodine removal	lodine mitigation measures implemented/ planned	Measurement of airborne iodine (gaseous and particulate form) activity in containment	Measurement of controlled iodine (gaseous and particulate form) release into environment	Other AM measures to mitigate airborne iodine (in gaseous/ particulate form) activity in containment (7)	Comments
all	none		airborne activity is measured but this does not separate iodine activity	The activity release to the environment is measured during activation of the filter		

A study of the long-term effects after a severe accident has been performed in Sweden both for BWR's and PWR's. One of the results from this project was that a pH in the range of 10-10.5 is desirable to minimize corrosion in the containment and also to reduce the iodine activity in the containment. Thus, a recommendation from this project was to adjust the activity in the containment by use of chemical additives very soon after the beginning of a severe accident in order to mitigate adverse long term-term effects. In Forsmark the work has continued and a study has been carried out which shows how a desirable level of pH can be achieved. Later on the intention is to implement suitable measures to add chemicals in the (short term) accident management to mitigate long-term effects.

Czech Republic:

Plant Name	Туре	Designer	Operational date	Thermal power (MWt)	Containment type	Free volume (m³)	Sprays	Containment Sump chemical Additives	Filters ¹⁾	Containment Venting filters
Dukonavy 4 units	VVER- 440/213	LOTEP Leningrad (former USSR), Energoprojekt Prague (Czech Republic)	1 st unit: March 1985 2 nd unit: January 1986 3 rd unit: December 1986 4 th Unit: July 1987	1375 (each)	Hermetic rooms surrounding primary loops are connected by the tunnel with the bubble condenser tower	gas volume: 51000 Pool volume: 1380	Three independent trains of containment sprays, flow capacity: 400 m³/h, additive: hydrazine (0.2gN ₂ H ₄ .H ₂ O+2.7g KOH+13.7gH ₃ BO ₃ per liter of spray solution)	no additives	- HEPA filters (different numbers in two different locations Capacity: 500 m³/h, DF=10³-10⁴ - active carbon filters (different numbers in two different locations Capacity: 1500 m³/h, DF=10 for I₂	no venting system

¹⁾ All these filters are used in normal operation conditions or refueling and maintenance; they will not be used in most types of accident conditions.

Plant Name	Passive iodine removal	lodine mitigation measures implemented/ planned	Measurement of airborne iodine (gaseous and particulate form) activity in containment	Measurement of controlled iodine (gaseous and particulate form) release into environment	Other AM measures to mitigate airborne iodine (in gaseous/ particulate form) activity in containment	Comments
Dukonavy 4 units	no intentional passive iodine removal	Hydrazine addition to the spray solutions (implemented)	Measurement of aerosol deposition on filters and of gaseous iodine	No measurement of controlled iodine release	No other dedicated AM measures	

Canada:

Plant Name	Туре	Designer	Operational Date	Thermal Power MW (t) per reactor	Containment Type	Free Volume (m³)	Sprays	Containment Sump Chemical Additives	Filters
Pickering A	CANDU	Ontario Hydro	1971	1744	Negative Pressure	3.15+5*	Dousing System in the Vacuum Building No chemical additives	none	The exhaust line of the normal ventilation system has a prefilter, HEPA filter and carbon filter
Pickering B	CANDU	Ontario Hydro	1982	1744	Negative Pressure	3.18+5*	Dousing System in the Vacuum Building No chemical additives	none	The exhaust line of the normal ventilation system has a prefilter, HEPA filter and carbon filter
Bruce A	CANDU	Ontario Hydro	1977	2832	Negative Pressure	1.59+5*	Dousing System in the Vacuum Building No chemical additives	none	The exhaust line of the normal ventilation system has a prefilter, HEPA filter and carbon filter
Bruce B	CANDU	Ontario Hydro	1984	2832	Negative Pressure	1.59+5*	Dousing System in the Vacuum Building No chemical additives	none	The exhaust line of the normal ventilation system has a prefilter, HEPA filter and carbon filter
Darlington	CANDU	Ontario Hydro	1990	2776	Negative Pressure	2.28+5*	Dousing System in the Vacuum Building No chemical additives	none	The exhaust line of the normal ventilation system has a prefilter, HEPA filter and carbon filter
Point Lepreau	CANDU 6	AECL	1982	2058		5.02+4	Design flow rate - 4530 kg/s Max flow rate - 6804 kg/s Hydrazine (100 mg/kg) No recirculation	none	Normal filtered air discharge system (FADS) includes a prefilter, two HEPA filters and a carbon filter
Gentilly 2	CANDU 6	AECL	1982	2058		5.02+4*	Design flow rate - 4530 kg/s Max flow rate - 6804 kg/s Hydrazine (100 mg/kg) No recirculation	none	Normal filtered air discharge system (FADS) includes a prefilter, two HEPA filters and a carbon filter

^{*} Total containment volume of four Reactor Buildings and the pressure relief system including the Vacuum Building

Plant Name	Containment Venting Filters	Passive lodine Removal	lodine Mitigation measures	Measurement of airborne iodine in containment	Measurement of controlled iodine (gaseous release into environment	Other AM measures to mitigate airborne iodine in containment ⁽¹⁾
Pickering A	Emergency Filtered Air Discharge System with two trains of filters, containing demisters, heaters, prefilters, two HEPA filters and carbon filters	none	none	lodine activity is measured the recirculation ventilation flow	lodine activity is continuously measured at the stack exhaust	
Pickering B	Emergency Filtered Air Discharge System with two trains of filters, containing demisters, heaters, prefilters, two HEPA filters and carbon filters	none	none	lodine activity is measured the recirculation ventilation flow	lodine activity is continuously measured at the stack exhaust	
Bruce A	Emergency Filtered Air Discharge System with two trains of filters, containing demisters, heaters, prefilters, two HEPA filters and carbon filters	none	none	lodine activity is measured the recirculation ventilation flow	lodine activity is continuously measured at the stack exhaust	
Bruce B	Emergency Filtered Air Discharge System with two trains of filters, containing demisters, heaters, prefilters, two HEPA filters and carbon filters	none	none	lodine activity is measured the recirculation ventilation flow	lodine activity is continuously measured at the stack exhaust	
Darlington	Emergency Filtered Air Discharge System with two trains of filters, containing demisters, heaters, prefilters, two HEPA filters and carbon filters	none	none	lodine activity is measured the recirculation ventilation flow	lodine activity is continuously measured at the stack exhaust	
Point Lepreau	Feasibility of using the FADS for post- accident venting is currently being evaluated	none	none	lodine activity is measured the recirculation ventilation flow	lodine activity is continuously measured at the stack exhaust	
Gentilly 2	No post-accident venting	none	none	lodine activity is measured the recirculation ventilation flow	lodine activity is continuously measured at the stack exhaust	

no information provided

The Netherlands:

Plant Name	Туре	Designer	Operational date	Thermal power (MWt)	Containment type	Free volume (m³)	Sprays	Containment Sump chemical Additives	Filters ¹	Containment Venting filters
Borssele	2-loop PWR	Siemens KWU	1972	1366	Steel and concrete	37100	Flow capacity: 100 m ³ /h, no additives	none	Active carbon filters	Venturi scrubber and metal filter, Venturi water: 7.0 m3 with 0.5 mass% NaOH and 0.2 mass% Na $_2$ S $_2$ O $_3$ Metal filter: 2 fases of DIN 1.4404 Aerosol absorbing capacity: >150 kg Efficiency - Large aerosols (1-2 mm): \geq 99.999% -Small aerosols (0.5 mm): \geq 99.998 % - Iodine (elemental): \geq 99.5 % - Iodine (organic): \geq 90% -Iodine resuspension (18 h): \rangle 0.1 % - Aerosol resuspension (18 h) 0.0034 %

Plant Name	Passive iodine removal	lodine mitigation measures implemented/ planned	Measurement of airborne iodine (gaseous and particulate form) activity in containment	Measurement of controlled iodine (gaseous and particulate form) release into environment	Other AM measures to mitigate airborne iodine (in gaseous/ particulate form) activity in containment	Comments
Borssele	none	spray system	yes	yes	none	

U.S.A

Plant name	Туре	Designer	Operational date	Thermal power (MWt)	Containment type	Free volume (1, 2)	Sprays (3)	Containment sump chemical additives (4)	Filters (5)	Containment venting filters (6)
Peach Bottom	BWR	GE	1974	3190	Mark I	1.59x10 ³	none	none	(9)	none
La Salle	BWR	GE	1982	2234	Mark II	1.85x10 ⁶	6700	none	(9)	none
Grand Gulf	BWR	GE	1983	3750	Mark IV	1.40x10 ⁶	5650	none	(9)	none
Surry	PWR	W	1972	2325	(6)	1.86Yx10 ⁶	960	pH>7.0 TSP (8)	none	none
Zion	PWR	BEW	1974	3150	(7)	2.72x10 ⁶	3000	pH>7.0 NaOH	none	none
TMI	PWR	BEW	1974	2400	(7)	2.16x10 ⁶	2500	pH>7.0 NaOH	none	none
Calvert Cliff	PWR	CE	1975	2535	(7)	2.10x10 ⁶	1400	pH>7.0 TSP (8)	none	none

- (1) Containment (gas volume ft³)
- (2) Drywell, Wetwell (gas/Pool volume m³
- (3) Flow capacity (injection phase/recirculation phase), chemical additive (if yes, what concentration), gpm
- (4) Chemical name, expected concentration, expected pH
- (5) Type (e.g., Prefilters, HEPA filters, active carbon filters), designed absorbing capacity and efficiency
- (6) Subatmospheric
- (7) Large dry containment
- (8) TSP stands for trisodiumphosphate
- (9) Standby gas treatment system (prefilters, HEPA and charcoal adsorbers, 95 to 99 % efficiency)

Plant Name	Passive iodine removal (10)	lodine mitigation measures implemented/ planned	Measurement of airborne iodine (gaseous and particulate form) activity in containment	Measurement of controlled iodine (gaseous and particulate form) release into environment	Other AM measures to mitigate airborne iodine (in gaseous/ particulate form) activity in containment (11)	Comments
Peach Bottom	none	none	yes	yes	none	
La Salle	none	none	yes	yes	none	
Grand Gulf	none	none	yes	yes	none	
Surry	none	none	yes	yes	none	
Zion	none	none	yes	yes	none	
TMI	none	none	yes	yes	none	
Calvert Cliff	none	none	yes	yes	none	

⁽¹⁰⁾ by traps and getters (e.g., special paints)
(11) in the stack during containment venting, speciation

Japan:

Plant Name	Туре	Designer	Operational date	Thermal power (MWt)	Containment type	Free volume (m³)	Sprays	Containment Sump chemical Additives	Filters ¹⁾	Containment Venting filters
Kashiwazaki Kariwa -3	BWR	Toshiba	11.8.1993	3293	Improved MARK-II	Containment gas volume: 14400 Drywell gas volume: 8700 Wetwell gas volume: 5700 Wetwell pool volume: 4000	Injection phase capacity: 0 m3/h Recirculation phase capacity: 1690 m3/h Chemical additive: No	none	- Prefilter - HEPA >99.97 % - Charcoal filter > 99.99 %	suppression pool scrubbing
Ohi - 3	PWR	Mitsubishi Heavy Industry	18.12.1991	3423	PCCV (1)	Containment gas volume: 73700	Injection phase capacity: 2400 m³/h Recirculation phase capacity: 2400 m³/h Chemical additive: Hydrazine (35 wt%)	NaOH (30 wt%) added through containment spray line Expected pH: 7	- Prefilter - HEPA > 99.% - Charcoal filter > 95 %	none

¹⁾ Prestressed Concrete Containment Vessel

Plant Name	Passive iodine removal	lodine mitigation measures implemented/ planned	Measurement of airborne iodine (gaseous and particulate form) activity in containment	Measurement of controlled iodine (gaseous and particulate form) release into environment	Other AM measures to mitigate airborne iodine (in gaseous/ particulate form) activity in containment	Comments
Kashiwazaki Kariwa -3	- Spray - SGTS (2) Filter - Suppression pool scrubbing in case of containment venting	PASS (3)		Yes	-	
Ohi - 3	- Spray - Filter	PASS		No venting in case of accident	-	

- 2) Standby gas treatment system
- 3) Post accident sampling system

Switzerland:

Plant Name	Туре	Designer	Opera- tional date	Thermal power (MWt)	Containment type	Free volume (m³)	Sprays	Containment Sump chemical Additives	Filters	Containment Venting filters
Beznau I + II	PWR (2 loops)	W	1969	1130	large dry	36'380	427'000 I/h in injection and recirculation phase borated water	no additives	no special filters for severe accidents	Sulzer-wet aerosol scrubber; additives: sodium carbonate and sodium thiosulfate; Capacity 5.2 kg/s of saturated steam; DF (Aerosol): minimum 1000 DF (elementary iodine): minimum 100
Leibstadt	BWR/6	GE	1984	3138	Mark III	Drywell: 7'768 Pool: 3'670 Wetwell: 36'183	none	no additives	no special filters for severe accidents	Sulzer-wet dust aerosol scrubber; additives: sodium carbonate and sodium thiosulfate; Filter load: max 150 kg; Capacity: 13.77 kg/s at 2.55 bar; DF (Aerosol): minimum 1000 DF (elementary iodine): minimum 100
Mühleberg	BWR/4	GE	1972	1097	Mark I	Drywell: 3'100 Torus pool: 2'200 Torus free volume: 2'050	above the drywell floor, 25 l/s, no additives	no additives	no special filters for severe accidents	Multi Venturi scrubber DF(Aerosol): 1000 DF (elementary iodine): 100
Gösgen	PWR (3 loops)	Siemens /KWU	1979	3002	large dry	55'000	none	no additives	no special filters for severe accidents	Venturi scrubber with integrated metal mesh filter. Capacity: 8.02 kg/s at 5.89 bar(a) (53 % Steam, 23 % air, 10 % H₂, 10 % CO₂, 4 % CO) Efficiency: - Aerosol (SnO₂) ≥ 99.99 % - Elemental iodine ≥ 99.5 %

Plant Name	Passive iodine removal	lodine mitigation measures implemented/ planned	Measurement of airborne iodine (gaseous and particulate form) activity in containment	Measurement of controlled iodine (gaseous and particulate form) release into environment	Other AM measures to mitigate airborne iodine (in gaseous/ particulate form) activity in containment	Comments
Beznau I + II	none	none	yes	yes	none	
Leibstadt	none	none	yes	yes	none	
Mühleberg	none	none	yes	yes	none	
Gösgen	none	none	yes	yes	none	

Germany

Plant Name	Туре	Designer	Operational date	Thermal power (MWt)	Containment type	Free volume (m³)	Sprays	Containment Sump chemical Additives	Filters	Containmen t Venting filters
Biblis A	PWR	KWU	1974	1225	Spherical steel	70000	100 m ³ /h boric water 3000 ppm	none (until now)		b); c)
Biblis B	PWR	KWU	1976	1300	Spherical steel	70000	2x70 m³/h boric water 3000 ppm	\		b); c)
Brokdorf	PWR	KWU	1986	1440	Spherical steel	70000	-			b); c)
Brunsbüttel	BWR	KWU	1976	806	Spherical steel	3816/2284*	-			a); b)
Emsland	PWR	KWU	1988	1363	Spherical steel	70000	-			b); c)
Grafenrheinfeld	PWR	KWU	1981	1345	Spherical steel	70000	-			a); b)
Grohnde	PWR	KWU	1984	1430	Spherical steel	70000	-			b); c)
Gundremmingen B	PWR	KWU	1984	1344	Concrete	8640/6068*	-			a); b)
Gundremmingen C	BWR	KWU	1984	1344	Concrete	8640/6068*	-			a); b)
Isar-1	BWR	KWU	1977	907	Spherical steel	3708/2252*	-			a); b)
Isar-2	PWR	KWU	1988	1420	Spherical steel	70000	-	\		a); b)
Krümmel	BWR	KWU	1983	1316	Spherical steel	4970/2714*	-			a); b)
Neckar-1	PWR	KWU	1976	840	spherical steel	52000	-			a); b)
Neckar-2	PWR	KWU	1989	1365	Spherical steel	70000	-			a); b)

Plant Name	Туре	Designer	Operational date	Thermal power (MWt)	Containment type	Free volume (m³)	Sprays	Containment Sump chemical Additives	Filters	Containme nt Venting filters
Obrigheim	PWR	KWU	1968	357	Spherical steel	35000	10-40 m³/h boric water 3000 ppm			a); b)
Philippsburg-1	BWR	KWU	1979	926	Spherical steel	3700/2172*	-			a); b)
Philippsburg-2	PWR	KWU	1984	1424	Spherical steel	72000	-			a); b)
Stade	PWR	KWU	1972	672	Spherical steel	45000	100 m³/h boric water 2200 ppm			b); c)
Unterweser	PWR	KWU	1978	1350	Spherical steel	70000	-			b); c)

Air volume in drywell/wetwell Venturi scrubber

- a)
- Metal fiber
- b) d) Molecular sieve

Plant Name	Passive iodine removal	lodine mitigation measures implemented / planned	Measurement of airborne iodine (gaseous and particulate form) activity in containment	Measurement of controlled iodine (gaseous and particulate form) release into environment	Other AM measures to mitigate airborne iodine (in gaseous/ particulate form) activity in containment	Comments
Biblis A			sampling system in preparation to be installed, mainly for PWRs			
Biblis B			↓			
Brokdorf						
Brunsbüttel						
Emsland						
Grafenrheinfeld						
Grohnde						
Gundremmingen B						
Grundremmingen C						
Isar-1						
Isar-2						
Krümmel						
Neckar-1						
Neckar-2						
Obrigheim						
Philippsburg-1						

Plant Name	Passive iodine removal	lodine mitigation measures implemented / planned	Measurement of airborne iodine (gaseous and particulate form) activity in containment	Measurement of controlled iodine (gaseous and particulate form) release into environment	Other AM measures to mitigate airborne iodine (in gaseous/ particulate form) activity in containment	Comments
Philippsburg-2						
Stade						
Unterweser			↑			