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Report of Foreign Travel of A. P. Malinauskas to England, Austria, and Germany

Foreign Trip Report

A. P. Malinauskas

September 9, 1982

Robert B. Minogue, Director Division of Reactor Safety Research

This document was prepared primarily for preliminary or internal use. It has not received full review and approval. Since there may be substantive changes, this document should not be considered final.

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INTERIM REPORT

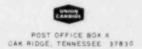
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ORNL FOREIGN TRIP REPORT

ORNL/FTR-1365

DATE:

September 13, 1982

SUBJECT: Report of Foreign Travel of A. P. Malinauskas, Head, Chemical Development Section, Chemical Technology

Division

TO:

Herman Postma

FROM:

A. P. Malinauskas

PURPOSE: To participate in the IAEA "Technical Committee Meeting on Fission Product Release to the Environment Following Class 9 Accidents," and to visit several European nuclear energy installations to exchange technical information on fission product chemistry, release, and transport in light water reactor accidents.

SITES VISITED: 8/20/1982

UKAERE Harwell, England P. Potter 8/23-27/1982 IAEA Vienna, Austria L. Epel 8/30/1982 KfK Karlsruhe, W. Germany H. Albrecht 8/31/1982 NUKEM Hanau, W. Germany H. Wingender

ABSTRACT: The traveler was a participant in the International Atomic Energy Agency Technical Committee Meeting on "Fission Product Release to the Environment Following Class 9 Accidents," which was held August 23-27, 1982, in Vienna, Austria. Prior to the meeting, the author visited the United Kingdom Atomic Energy Research Establishment at Harwell, and after the meeting visited Kernforschungszentrum Karlsruhe, and NUKEM GmbH, Hanau, West Germany. Discussions at these sites, as at the IAEA meeting, primarily involved fission product release, chemistry, and transport during light water reactor accidents. This document is a report of these discussions.

REPORT OF FOREIGN TRAVEL OF

A. P. MALINAUSKAS

Head, Chemical Development Section, Chemical Technology Division, ORNL

I. Introduction

The primary purpose of this trip was to participate in the International Atomic Energy Agency (IAEA) Technical Committee Meeting on "Fission Product Release to the Environment Following Class 9 Accidents," which was held August 23-27, 1982 in Vienna, Austria. At that meeting, I reviewed current work being conducted [primarily under Nuclear Regulatory Commission (NRC) sponsorship] in the United States in the area of fission product release from fuel and its subsequent behavior in severe core damage accidents.

In addition, site visits were made to the Atomic Energy Research Establishment (AERE), Harwell, United Kingdom, and to the Kernforschungszentrum Karlsruhe (KfK), West Germany, for purposes of technical exchange on fission product release, chemistry, and transport in light water reactor (LWR) systems. A site visit was also made to NUKEM GmbH, Hanau, West Germany, where discussions of fission product chemistry were extended to include waste management and decontamination considerations.

II. Site Visits

As already indicated, three sites were visited either prior to or immediately after the IAEA Technical Committee Meeting. Each of these site visits is described individually in this section.

Atomic Energy Research Establishment, Harwell

AERE Harwell was visited August 20, 1982. In the morning, round-table discussions were held with those persons in attendance who are listed in Appendix A, "Persons Contacted," of this report, whereas the agenda in the afternoon involved private discussions with P. Potter of Harwell and R. Hargreaves of the United Kingdom Atomic Energy Authority (UKAEA) facility at Windscale. These two were the principal authors of the documentation on fission product chemistry and behavior, which is being readied in preparation for the Public Inquiry concerning the Sizewell B Power Station. (Discussions over an extended lunch period were also held with R. Flowers, who did not participate in the morning meeting.)

Considerable interest was expressed in activities concerning the Three Mile Island (TMI) reactor; this interest ranged from the possibility of deriving further inferences regarding fission product behavior during the TMI accident to what was observed in the "Quick Look" tests (which involved the insertion of a closed-circuit television camera into the reactor vessel). The UKAEA had been sent videotapes of the Quick Look tests, but it was unclear whether these tapes were sent from NRC or

4

from the Electric Power Research Institute (EPRI). (Amusingly, the tapes were sent to the Information Center. The staff there had no idea what the tapes were, nor who they were for!)

Little experimental work has been done on fission product release and chemistry in the United Kingdom thus far, as most of the effort has been expended in developing expertise in this area through careful study of the literature and in preparing for the upcoming Public Inquiry.

Preparations are being made by Nichols (who was not present at Harwell) to investigate fission product release from LWR fuel. Morever, some work has been initiated on studies of the effects of radiation on iodine aqueous chemistry. The results obtained to date suggest that the radiation effects are small. This is in conflict with results reported by M. Lucas et al. in France. Attempts to duplicate the French data have not been succssful; the British results indicate 10-100 times less iodine production (as 1_3^-) in iodide solutions than the French data.

Studies have also begun which are directed toward modeling photochemical reactions of iodine. Experiments have been conducted which indicate that iodine and ozone will undergo a photochemical reaction to produce an iodo-oxy radical. This radical, in turn, polymerizes at a rate significantly more rapid than that characteristic of a simple nucleation process to form a submicron-size aerosol, of apparent chemical formula I409. It should be noted, however, that the tests were conducted in a moisture-free atmosphere, and it is not clear how these results will be altered by the presence of water vapor. Attempts will also be made to produce and characterize HOI in the gas phase; if successful, the gaseous species will be contacted by water in an effort to determine the corresponding partition coefficient.

Some in-pile experiments with Advanced, Gas-Cooled Reactor (AGR) uranium dioxide fuel were described in which the experimenters claimed that, when the fuel pin was effectively purged with an oxidizing gas (CO₂), the iodine and noble gas releases were virtually identical, whereas when a fuel pin was ruptured, with no opportunity for CO₂ ingress into the pin, the iodine releases were at least two orders of magnitude less than that of the noble gases. From this they concluded that the gap inventories in AGR fuel pins (presumably of the long-lived isotopes and expressed as fraction of total inventory) were identical for fission product iodine and the noble gases, but that the iodine was present in a chemically combined form.

Kernforschungszentrum Karlsruhe

The agenda at KfK consisted of a number of private discussions with those listed in Appendix A of this report and an impromptu seminar which I presented late in the afternoon of August 30 to 20 to 30 staff members. The primary subject of the seminar concerned the status of recovery efforts at TMI, but a significant part of the discussion after the "formal" presentation concerned fission product chemistry and transport.

The studies of fission product releases under core-melt conditions which were being conducted by Albrecht and Wild in their SASCHA facility have been concluded. This is partly by design and partly in response to criticism concerning the applicability of simulated irradiated fuel which they employed in their work. Criticism has been particularly severe regarding the results corresponding to releases of the more volatile species (notably cesium, iodine, and tellurium) and the results at lower temperatures (less than about 2000°C) in general.

Consequently, the SASCHA facility is currently being used to investigate core-concrete interactions and fission product release and aerosol formation resulting therefrom.

Because of difficulties in preparing and characterizing HOI, the doctoral research of Mr. Borkowski, which was initiated about one year ago, has been changed to involve studies of the formation and decomposition of methyl iodide, CH3I, as applied to LWR accidents. The HOI studies have not been completely abandoned, however; these have been undertaken by Dr. Hahn. His approach is similar to one being pursued by Dr. Cox at Harwell. Both will attempt to prepare HOI by a matrix isolation technique as was done some years ago at a temperature of about 5 K at the U.S. Naval Research Laboratory.

Mr. Wilhelm related that one of the more interesting results of the updated German Reactor Safety Study concerned the analysis of a core-melt accident with containment failure some four days after the meltdown. He stated that, for such an accident, although the release of fission product iodine from the core would be quantitative, the analyses indicate less than 10 Ci of this fission product would actually enter the biosphere. Mr. Wilhelm was extremely skeptical of the result.

I had hoped to discuss with Peter Hosemann his model for iodine behavior in an LWR accident. Unfortunately, he was in the U.S. at the time, but indicated a desire to discuss the model with me during the upcoming NRC Reactor Safety Research Information Meeting in Gaithersburg. There was noticeable reluctance to discuss the model at KfK; indication was made, however, that the model does not explicitly consider iodine chemistry.

NUKEM

The agenda for the August 31 visit to NUKEM, Hanau, West Germany, began with a round-table discussion involving virtually all of the individuals identified in Appendix A, followed by meetings with them individually throughout the remainder of the day. Much of the discussions, while interesting and informative, touched only peripherally on the area of reactor safety. The NUKEM activities which were described to me were primarily in the areas of waste volume reduction, waste management, decontamination and sealing, and spent fuel storage and transport. Their interest in our safety studies evolves primarily from the spent fuel storage and transport involvements.

Appendix D, Reports Received, identifies documents which I received at NUKEM and which adequately describe most of the information that was transmitted in the course of the discussion. Copies of these documents can be made available to requesting individuals.

Two items that are not included in the documents identified in Appendix D, but that were discussed, include the operation of a Thermic Volume Reduction Pilot Plant and a model for aerosol production involving aqueous systems. I was given a tour of the Thermic Volume Reduction Pilot Plant, which was operating at the time.

Thermic Volume Reduction is a scheme by which contaminated combustible waste is first gasified at elevated temperatures in a steam-air atmosphere and the gaseous effluent from the gasifier is subsequently combusted in a second furnace in a propane-oxygen-steam atmosphere. (Two-stage pyrolysis is employed in order to obtain an easily recoverable actinide product from the resultant ash.) The unit had been successfully operated for approximately two years with noncontaminated waste. Uranium, as a uranyl nitrate solution, was to be added the next day.

The aerosol formation modeling studies are directed toward aerosol transport problems that have been identified with spent fuel storage pools and with fuel reprocessing operations. Nonetheless, the model, which is being developed by H.-J. Dorst, may be useful in assessing effects arising from the flashing or less violent evaporation of fission product-containing aqueous pools. In essence, the model accepts a clathrate structure of an aqueous solution and presumes more or less uniform "evaporation" of the system. The aerosol is then regarded as consisting initially of macromolecules of the aqueous system.

Dorst claims good correlation between the model and Nishima's studies at Hanford concerning aerosol formation above radioactive liquids under conditions of evaporation, nucleate boiling, and air injection. Experiments at NUKEM are planned to critically test the model.

III. IAEA Technical Committee Meeting on "Fission Product Release to the Environment Following Class 9 Accidents"

This meeting was held August 23-27 in the Vienna International Center, Vienna, Austria. The list of attendees is presented in Appendix B, and the agenda is listed in Appendix C. In addition, on Tuesday, August 24, I also met with Mr. Blix, the Director General of IAEA, to describe and display excerpts from the television tapes of the TMI "Quick Look" tests that were brought to the meeting by Dr. Pasedag of the NRC.

Mr. E. V. Gilby, who is chairman of the committee, described the meeting during his closing remarks as "unexpectedly informative." This is an apt description, as many felt the meeting was premature in terms of technical progress since the previous meeting, and with no clearly stated objective.

Indeed, little technical progress had been achieved since the first meeting; this was made especially obvious in updating the previous report of the committee. In important technical areas, the only modifications to the earlier text that were suggested concerned recognition that work had been initiated in the period between the two meetings.

The primary objective of the first meeting was, as the committee title implies, to develop a consistent body of fission product source terms for use in analyses of severe core damage accidents. With the explosion of activity in this area by the Organization of European Community Development/Nuclear Energy Agency, the American Nuclear Society, and the individual German, British, Dutch, and EPRI reactor safety studies, as examples, the stated objective is redundant, to say the least. It is therefore likely that the purpose of the committee will be modified to emphasize regulatory aspects to a greater extent.

The ill-defined purpose of this meeting was almost unanimously recognized. As a consequence, a Consultant's Meeting will be held prior to any future meetings of the Technical Committee in order to focus the meeting and to determine its timing relative to a number of anticipated forthcoming developments.

As with the previous meeting, the number of Member States participating is disappointingly small. It is particularly disturbing to note that Germany, a previous participant, saw fit not to send a representative. Unless participation of Member States is expanded, the United States should seriously reconsider its own participation.

A number of interesting views were presented in the course of the meeting. Some of these are presented below, in approximately chronological order, with little or no elaboration:

- The Argentinian representative indicated that his country was primarily concerned with reactor accident effects on land impaction, so that iodine releases were of lesser concern than the releases of longer-lived nuclides (e.g., cesium). In Austria, hydrogen explosions were of great concern.
- A number of delegations (Czechoslovakia, the Netherlands, the United Kingdom) expressed the need for realistic, as opposed to conservative, approaches to consequence analysis, particularly where application involves emergency planning.
- The Central Electricity Generating Board (CEGB) of the United Kingdom has claimed it has designed away V sequence accidents (accidents involving containment bypass) in their plants. The U.K. Regulatory member of the delegation quickly noted, however, that the claim is under review and has not yet been accepted by their licensing body.
- The Marviken multinational aerosol test program in Sweden appears to be receiving strong support from code developers. I have a number of serious concerns about the program, however, and voiced two of these:

- (1) Why can no thermal-hydaulic code be suitably modified so it too can be tested? That is, why is it necessary to input directly-measured thermal-hydraulic parameters into the aerosol code? (2) Are the aerosol code input parameters (other than temperatures and flow rates) all known a priori, or will it be necessary to adjust the parameters to fit the experimental data? If adjustment is necessary, then Marviken is an elaborate and expensive experiment which will merely demonstrate a code's ability to correlate data a trivial exercise in view of the number of parameters that can be adjusted.
- The United Kingdom risk analyses have introduced a new concept with respect to source term assessment, the "discrete probability distribution" (DPD), which is a measure of the probability that a particular source term point value is indeed this value. The distribution function itself is developed by having a group of "experts" essentially vote on the matter. I could not help but remark that the U.K. approach has introduced a new level of obfuscation into risk analysis.
- SUPER-SARA, a multinational program at Ispra, Italy, has as its objectives the determination of the extent of core damage and flow blockage due to transients, and the extent of fission product release during the transients. The initial experiments, which are scheduled for 1985, are to examine large break loss-of-coolant accidents and will not evaluate fission product behavior. Subsequent tests, which include fission product monitoring, will be conducted in three series: (1) clad rupture tests (1100-1650 K), (2) rubble bed formation tests (1900 K), and (3) tests resulting in molten material (2300 K). There is serious concern regarding the timeliness of the entire operation, particularly in view of limited progress to date and escalating costs; a decision on whether or not to proceed will made in autumn 1982.

IV. Concluding Remarks

The utility of site visits as a means of maintaining active technical exchanges among researchers in the field is unquestioned. I can only continue to applaud the extent to which NRC supports international technical exchange in the area of reactor safety research.

The utility of the IAEA Technical Committee meetings, on the other hand, should be more carefully scrutinized. I can see great value if the meeting is one primarily involving regulators, with technical members present to apprise the regulators of the state of the technology and to ensure that technical soundness is maintained. As a technical exchange, however, it is superfluous. Moreover, unless the meeting is more fully supported by the IAEA Member States, continued U.S. participation should be reexamined.

Appendix A

Persons Contacted

UKAERE Harwell

John Amphlett - Chemistry Division, Harwell W. G. Burns - Chemistry Division, Harwell

David Clough - Harwell

Tony Cox - EMS Division, Harwell

M. Deane - Chemistry Division, Harwell

Alan Eggleton - EMS Division, Harwell

Ron Flowers - Fuel Recycle Program, Harwell

Ron Hargreaves - UKAEA Windscale

Norman Large - Chemistry Division, Harwell

G. Low - UKAEA Directorate

Paul Potter - Chemistry Division, Harwell John Sayers - Metallurgy Division, Harwell

Jack Williams - UKAEA Directorate

Kernforschungszentrum Karlsruhe

H. J. Ache Helmut Albrecht Borkowski Hahn Hans Wild J. Wilhelm

NUKEM

H.-J. Dorst - Safety Technology Division

Jürgen Hofmann

R. Leicht - Safety Technology Division

D. Neupert - Research and Development Section

E. Pollmann - Transnuclear

E. Schlich

H. J. Wingender - Safety Technology Division

H. Witte - Research and Development Division

11

Appendix B

List of Participants, IAEA Technical Committee Meeting on Fission Product Release to the Environment Following Class 9 Accidents

Department of Nuclear Energy and Safety Division of Nuclear Safety	nd Safety		Lasue no. 2 1982-08-25
	NOTIFICATION OF AN AGENCY SPONSORED MEETING	ORED MEETING	
Subject of Meeting : Technical Environment	Technical Committee Meeting on Fission Product Release to the Environment Following Class 9 Accidents	o the	Responsible Officer : Lester Epel Re-w A2617 Ext. 2694/2697
Dates : 23 - 27 A	27 August 1982		
Place : IAEA, Hea	Headquarters, Meeting Room IV, ext. 1341		Opening : 10 a.m.
PARTICIPANTS AND DESIGNATING NEMBER STATES AND ORCANIZATIONS	Abroad	In Vienna	FOR THE PERIOD
ARCENTINA			
Mr. P. Sentason	Argentine AEC Avenida Libertador 8250 Buenos Aires 1429	Hotel Astoria Kaertnerstr. 32 1010 Vienna	23 - 27 August 1982
AUSTRIA			
Mr. Nilos Koaurka	Oesterreichisches Forschungszentrum Selbersdorf Ges.m.b.H Selbersdorf	Gumplowiczstr. 1 1220 Vienna	
CANADA			
Mr. D. Yren	Atomic Energy of Canada Ltd. Whiteshell Ruclear Research Establishment Plnawa, Manitoba ROE 110	Pension viener Seilergasse 16 1010 Vienna	
CZECHOSLOVAKTA			
Mr. 2. Kriz	Czechoslovak Atomic Energy Commission Department of Nuclear Safety & Safeguarda Slezska 9 12029 Frague 2	Pension Di vi Singergasse 8 1010 Vienna	
FINLAND			
Mr. Lelf Blomquist	Institute of Radiation Protection P.O. Box 268 SP-0v101 Helsinki 10	Hotel Am Stefansplatz Stephansplatz 9 1010 Vienna	latz
FRANCE			
Mr. Christian bevillers	Chef du service d'analyse et d'evaluation des risques Departement de surete nucleaire 8.P. 6 92260 Fontenay-aux-Roses	Horel Tigra Tiefergraben 14 1010 Vienna	23 - 27 August 1982

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PARTICIPANTS AND	ADDRESSES		FOR THE PERIOD
DESIGNATING MEMBER STATES AND ORGANIZATIONS	Abroad	In Vienna	
NDIA			
	Safety Research Laboratory	Hotel Elite	
Mr. A. R. Sunderarajan	Reactor Research Centre	Wipplinger Strasse 32	A . 18. 6
	Kalpakkam - 603102	1010 Vienna	
	Tamil Nadu		
TALY			
Mrs. C. Brofferio	CNEN CONTRACTOR DESCRIPTION	Hotel /.storia	1 452
	Nuclear Safety Division	Kaertnerstr. 32	
	Viale Regina Margherita 125 Rome	1010 Vienna	
	NOME.	1010 713000	
Mr. P. Cagnetti	CNEN		
	Nuclear Safety Division		
	Viale Regino Margherita 125		
	Roac		
Mr. Monilla	CNEN		
	Nuclear Safety Division		
	Viale Regina Margherita 125		
	Rome		
NETHERLANDS			
Mr. P.J. de Munk	Ministry of Social Affairs	Hotel Madeleine	
	Nuclear Department	Geblergasse 21	
	P.O. Box 69	1170 Vienna	The state of the s
	2270 MA Voorburg		17, 24
SPAIN			
Mr. F. Diaz de la Cruz	Radiological Protection Division	Hotel Kongress	
	Junta de Energia Nuclear	Wiednerguertel 34	1,5
	Avenida Complutense	1040 Vienna	
	Hadrid-3		
SWEDEN			
Mr. H. Hedgran	Department of Reactor Technology	Hotel Am Stephanplatz	
	Royal Institute of Technology		
	1044 Stockholm		
Mr. L. Devell	Studsvik Energiteknik AB	Hotel Urania	
m. a. bereit	Fack	Weissgerberstr. 7	
	S-611 82 Nykoping Sweden	1030 Vienna	

ARTICIPANTS AND	ADDRESSES	1 - 127 - 1	FOR THE PERIOD
SIGNATING MEMBER STATES ID ORGANIZATIONS	Abroad	In Vienna	
WITZERLAND			
Mr. S. Chakraborty	Division pour la Securite des	Hotel Nordbahn	
m. s. character	Instaliations nucleaires	Prateratr. 72	7.0
	CH-5303 Wuerenlingen	1020 Vienna	
NITED KINGDOM	Ch 3303 wherenitingen	1020 Vienna	
Mr. F. Abbey	UK AEA Safety & Reliability Directorate	Pension Schneider	
	Wigshaw Lane	Getreidemarkt 5	
	Culcheth, Warrington	1060 Vienna	
	WA3 4NE		
Mr. D.M. Bruce	Nuclear Installations Inspectorate	Hotel Kaertnerhof	
	Thames House North		11.5
		Grashofgasse 4	
	Hillbank, London SWIP 4QJ	1010 Vienna	
Mr. P.H. Cowley	Ministry of Defence (PE)	Pension Schneider	
	Block F		
	Foxhill, Bath		
Mr. R.L. Faircloth	UKAEA HATWELL		
HI. R.L. PAIRCIGE		Camping Site Haxenburg	
	Oxfordshire OXII ORA		
Mr. E.V. Gliby	Nuclear Safety Technology & Overseas	Pension Schneider	
(Chairman)	Collaboration		
	Safety & Reliability Directorate of the		
	UKAEA		
	Wigshaw Lane		
	Colcheth, Warrington WA34NE		
Mr. Sansom	Rolls-Royce and Associates Limited	Pension Schneider	
	P.O. Box 31	1.0000000000000000000000000000000000000	
	Derby DEZ 8BJ		
BITED STATES OF AMERICA			
NITED STATES OF AMERICA			
Mr. P. Baybutt	Battelle Columbus Laboratories	Pension Wicner	
	505 King Avenue		W
	Columbus, Ohio 43201		
Mr. R. Burus	EDS Nuclear Corporation	Harat Mile-	
III . II . Dat III	350 Lennon Lane	Hotel Hilton	
		Am Stadtpark	
	Walnut Creek, California 94598	i030 Vienna	
Mr. A.P. Hallmauskas	Chemical Technology Division, O.R.N.L.	Pension Wiener	
	P.O. Box X	The state of the s	*
	Oak Ridge, Tennessee 37830		
Mr. O. Michael	Provide the form of the first o		
Mr. H. Michael	Stone & Webster Engineering Corp.	c/o Dr. Gerhard	
	245 Summer Street	Landskrongasse 2/14	
	Boston, Mass. 02107	1010 Vienna	

PARTICIPANTS AND	ADDRESSES		FOR THE PERIOD
DESIGNATING MEMBER STATES AND ORGANIZATIONS	Abroad	In Vienna	
Mr. W. Pasedag	U.S. Nuclear Regulatory Commission Washington, D.C. 20555	Hotel Tigra	
Mr. W. R. Stratton	2 Acoma Lane Los Alamos NM 87543	Peter Jordanstrasse 67a/3 1180 Vienna	
NTERNATIONAL ORGANIZATIONS			
Commission of the European	Communities		
Mr. V.E. Della Loggia	Direction Generale de la Science 200 Rue de la Loi B-1049 Brussels, Belgium	Hotel Wiedner Staatsoper Krugerstrasse 11 1010 Vienna	
Mr. A. Schuerenkaemper	Joint Research Centre 21020 Ispia (Italy	Hotel Atlas Lerchenfelderstr.l Vienna	
DBSERVERS			
Mr. M. Hamard	Department de protection Centre d'etudes nuclealies BP 6 92260 Fontenay-aux-Roses France		
Mr. R. Vogel	Electric Power Research Institute P.O. Box 10412 Palo Alto, CA 94303 USA	Hotel Astoria	

17

Appendix C

Agenda of the IAEA Technical Committee Meeting on Fission Product Release to the Environment Following Class 9 Accidents

18

TECHNICAL COMMITTEE MEETING

ON

FISSION PRODUCT RELEASE TO THE ENVIRONMENT FOLLOWING CLASS 9 ACCIDENTS

23 - 27 August 1982

TINAL AGENDA

Monday	10:00 - 10:15	Introduction of Participants and Announcements	Scientific Secretary
	10:15 - 10:30	Welcome	Director, NENS
	10:30 - 11:00	Review and Revision of Agenda	Chairman
	11:00 - 11:20	COFFEE BREAK	
	11:20 - 11:50	Overview of ANS "white paper" of the Source Term	Stratton
	11:50 - 12:30	Discussion	
		LUNCH	
	14:15 - 14:45	Progress Report by Chairman of OECD-NEA Committee on the Source Term	Abbey
	14:45 - 15:00	Discussion	
	15:00 - 15:20	Review of FLASH Experiments	Devillers
	15:20 - 15:35	Discussion	
	15:35 - 16:00	COFFEE BREAK	
	16:00 - 17:00	Video-tape of TMI Core	Fasedag, Malinauskas
	13:00	WINE AND CHEESE PARTY	
Tuesday	09:15 - 09:45	Review of Current Problems on 2.7. Release from Fuel	Malinauskas
	09:45 - 10:30	Discussion	
	10:30 - 10:45	COFFEE BREAK	
	10:45 - 11:15	Review of Current Status of Experiments on Water Chemistry	Wren
	11:15 - 12:00	Discussion	
		LUNCH	

- 2 -

Tuesday Contd.	14:00 = 14:30	Status of Computer Code Development and Future Requirements	Baybutt
	14:30 - 15:00	Discussion	
	15:00 - 15:30	Marviken Aerosol Transport Tests	Devell
	15:30 - 16:00	Discussion	
	16:00 - 16:15	CCFFEE BREAK	
	16:15 - 16:45	Overview of NRC Sponsored Research on the Source Term	Pasedag
	16:45 - 17:15	Discussion	
Wednesday	09:00 - 09:15	Review of Knowledge of Aerosol Behaviour	Sundararajan
	09:15 - 09:30	Discussion	
	09:30 - 10:00	Update of Current Licensing Procedures and Regulatory Perspective following Revised NUREG-0771	Pasedag
	10:00 - 10:30	Discussion	
	10:30 - 11:00	COFFEE BREAK	
	11:00 - 11:30	Some Results on Icdine Chemistry in the Containment	Devillers
	11:30 - 12:00	Discussion	
		LUNCH	
	14:00 - 17:00	Round Table Review of Current National Positions *	
Thursday	09:00 - 10:30	Completion of Round Table Discussion	
	10:30 - 10:45	COFFEE BREAK	
	10:45 - 12:00	Draft Outline of Summary Report to the Agency	
		LUNCH	
	14:00 - 17:00	Draft Summary Report	
Friday	Morning	Review Draft and Make Changes	
		CLOSING OF THE MEETING	

Delegates are invited to give a brief overview of their country's regulatory position and projections of future trends vis-à-vis the source term. Coffee break as appropriate.

20

Appendix D

Reports Received

UKAEA, Harwell

Chemical Data for the Calculation of Fission Product Releases in Design Basis Faults in PWRs, S. M. Ali, R. J. Bawden, A. M. Deane, K. Garbett, and N. R. Large, AERE-R 10494 (March 1982).

The Analysis of Fission Products in the Windscale A.G.R. Coolant Using On-Line γ Spectrometry, R. Hargreaves and E. H. Perrott.

KfK

Untersuchung der Freisetzung von Spalt- und Aktivierungsprodukten beim Kernschmelzen - Fortschrittsbericht 1981, H. Albrecht and H. Wild, April 1982.

NUKEM

Brochures:

Mobile Waste Conditioning Plant, MOWA

Two-Stage Evaporator

Pyrolysis Plant for Volume Reduction

Liquid Waste Concentration System

Sealing Radioactively Contaminated Components

Peelable Decontamination Films

Transport/Storage Cask TN 1300 Technical Description

IAEA Technical Committee Meeting

Report of the Technical Committee Meeting on Thermal Reactor Safety Research, 1-4 December 1981, Moscow, USSR.

"The Marviken Aerosol Transport Tests (ATT)," L. Devell presentation to IAEA Technical Committee Meeting on Fission Product Release to the Environment Following Class 9 Accidents, Vienna, Aug. 23-27, 1982.

Description of Iodine Chemistry Studies at AECL, Whiteshell, Canada.

"Iodine Behavior in PWR Accidents Leading to Severe Core Damage,"
M. Lucas, C. Devillers, J. Fermandjian, and D. Manesse, preprint of paper
presented at ANS Topical Meeting on Thermal Reactor Safety, August 30,
1982, Chicago.

"Sizewell "B" Power Station Public Inquiry: Appendix M: Degraded Core Analysis."

"Design Considerations for Implementing a Vent-Filter System at the Barseback Nuclear Power Plant," J. Johansson, L. Nilsson, and Å. Persson, International Meeting on Thermal Nuclear Reactor Safety, Chicago, Illinois, August 29-September 2, 1982.

Report of Technical Committee Meeting on Airborne Fission Product Release Following Extensive Core Damage Accidents, October 12-16, 1981.

Report on IAEA Specialists Meeting on Early Diagnosis of Failures in Primary System Components of Nuclear Power Plants, 21-25 June 1981, Prague, CSSR.

Report on Technical Committee Meeting on Thermal Reactor Safety Research, 1-4 December 1981, Moscow, USSR.

DISTRIBUTION

- 1-2. Assistant Secretary for International Affairs, DOE, Washington
- 3. R. B. Minogue, Director, Office of Nuclear Regulatory Research, NRC, Washington
- 4. Director, Division of Safeguards and Security, DOE, Washington
- 5-6. Director, Division of International Security Affairs, DOE, Washington
- 7-8. Director, International Programs, NRC, Washington
- 9-10. Director, Division of Technical Information and Document Control, NRC, Washington
 - 11. Director, Division of Accident Evaluation, NRC, Washington
 - 12. J. A. Lenhard, DOE/ORO
 - 13. J. S. Denton, DOE/ORO
 - 14 Herman Postma
- 15-16. A. P. Malinauskas
 - 17. IETA Program Manager, Lawrence Livermore Laboratory, Mail Stop L-389, P. O. Box 808, Livermore, CA 94550
 - 18. M. Silberberg, Division of Accident Evaluation, NRC, Washington
 - 19. R. R. Sherry, Division of Accident Evaluation, NRC, Washington
 - 20. J. Larkins, Division of Accident Evaluation, NRC, Washington
 - 21. R. W. Houston, Division of Systems Integration, NRC, Washington
 - 22. C. N. Kelber, Office of Nuclear Regulatory Research, NRC, Washington
 - 23. J. T. Bell
 - 24. D. O. Campbell
 - 25. D. E. Ferguson
 - 26. T. S. Kress
 - 27. R. A. Lorenz
 - 28. A. L. Lotts
 - 29. D. B. Trauger
 - 30. R. P. Wichner
 - 31. R. G. Wymer
- 32-33. Technical Information Center, DOE, Oak Ridge
- 34-35. Laboratory Records Department
 - 36. Laboratory Records Department-RC
 - 37. Laboratory Protection Division
 - 38. ORNL Patent Office
 - 39. ORNL Public Relations Office