RECEIVED BY DTIE SEP 20 1967

MASTER

OAK RIDGE NATIONAL LABORATORY

operated by

UNION CARBIDE CORPORATION

NUCLEAR DIVISION

for the

U.S. ATOMIC ENERGY COMMISSION

ORNL - TM - 1899

COPY NO. - 66

DATE - June 21, 1967

CONF-660907--12

**DISTRIBUTION OF FISSION PRODUCTS FOLLOWING
A NUCLEAR INCIDENT**

Wm. B. Cottrell

ABSTRACT

The data on fission product release from actual reactor incidents and simulated accidents was reviewed. From the former it was evident that the measured releases, generally noble gas, were but a fraction of that for which the systems had been designed and that the consequent radiation injuries even to "radiation workers" constitute a very small fraction of their total injuries. The extensive research and development sponsored by the AEC on fission product behavior under simulated accident conditions has resulted in an understanding of the release mechanism as a function of many accident parameters. Furthermore, since consequence-limiting safety features are more demonstrable and acceptable than accident-limiting safeguards, the thrust of the R&D program is toward the transport and removal of released fission products and the verification of this behavior in large scale experiments.

NOTICE This document contains information of a preliminary nature and was prepared primarily for internal use at the Oak Ridge National Laboratory. It is subject to revision or correction and therefore does not represent a final report.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Reg

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

- A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or
- B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

FIG 300, MN 65

DISTRIBUTION OF FISSION PRODUCTS FOLLOWING
A NUCLEAR INCIDENT

by

Wm. B. Cottrell

A paper Prepared for Presentation
at the
152 National Meeting
of the
American Chemical Society

New York City
September 12-16, 1966

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

CONTENTS

	<u>Page</u>
Introduction	4
Relevant Commission Research and Development	5
The ORNL Research and Development Program	8
Fission Product Release in Actual Incidents	26
Discussion and Conclusions	35

INTRODUCTION

Since the inception of the nuclear energy program, those responsible for safety have been concerned about the release and distribution of fission products following a nuclear incident. In the early days of nuclear energy, the concern was of less public significance, since the reactors were small and their locations remote. However, existing power reactors are now pushing 1,000,000 kw thermal in a single unit, and other plants already contracted for, will be several times as large. Furthermore, locations in remote areas are not compatible with today's need to provide electrical power for our industrial complexes, since this need involves installed capacity near heavily populated areas. We are therefore, now, more than ever before, concerned with the potential release and distribution of fission products following credible reactor accidents.

When Dick Vogel, our Symposium Chairman, conceived of this program, he not only had this background in mind, but also was well aware of the role that chemical technology has, and is playing, in identifying and resolving the questions associated with the behavior of fission products in nuclear incidents. I was honored in that he asked me to present a survey paper on the subject and to serve as chairman of this morning's meeting. I have based this paper on the activities of the AEC's Nuclear Safety Research and Development program which is sponsored by the Commission's Division of Reactor Development and Technology, and will also touch lightly on the work of my colleagues, at ORNL and elsewhere, several of whom are presenting papers here also. In my paper this morning, I shall discuss briefly the Commission programs relating to the release and transport of fission products in simulated nuclear accidents and then summarize at greater length, the relevant ORNL activities. Following that, and in order to present this overall problem in its proper perspective, we will then consider briefly the experience in actual reactor incidents, both in terms of the fission products released and also in terms of the resulting exposure to individuals, and then reflect on where we seem to be headed.

RELEVANT COMMISSION RESEARCH AND DEVELOPMENT

The Commission's overall R&D program on Nuclear Safety¹ is, of course, much more comprehensive than that represented by our subject today. Thus, of the 24 million dollars expended on the Nuclear Safety Program in FY 66, I would estimate that around 4 million was spent on studies which are directly concerned here, not including some related studies in the space safety program. In addition to such direct expenditures, a somewhat larger amount of money per year goes into multipurpose projects such as LOFT and CSE, wherein the behavior of the fission products is but one aspect of the phenomenology in question. For those not familiar with the terminology, LOFT is the designation of the 50 Mw(t), PWR reactor which will be permitted to have a core meltdown at Nation Reactor Testing Station; CSE is the acronym for the Containment Systems Experiment at Hanford.

Research projects which bear on fission product behavior are many and varied, and include studies applicable to the three major reactor types, 1) water reactors (both PWR and BWR), 2) gas-cooled reactors, and 3) fast sodium reactors. It would be inappropriate for me to attempt to discuss each of these tasks, but in order that you might quickly have some comprehension of the number, nature, sponsoring organization, and level of effort, I have tabulated these tasks by reactor type in the next three tables. Table 1 lists the 13 tasks in this area being conducted for the water reactor program, Table 2 the 5 tasks in the gas reactor program, and Table 3 the 5 tasks in the fast reactor program.*

These three tables tabulate some 23 projects underway at some of eight major research organizations as of the end of last fiscal year. A few of these projects may be terminating and others may be initiated in FY 67, but by and large, these tables summarize the current research picture. They do not, however, include such research work as may be involved in some of the larger multipurpose projects as previously mentioned.

*Data from these tables is taken from Reference 1.

Table 1. Water Reactor Program
Research Tasks Relating to Fission Product Behavior

No.	Title	Sponsor	\$ in FY 66
1.	Behavior of Fission Products on the In-Pile Destruction of Reactor Fuels	ORNL	4-500,000
2.	Release of Fission Products under Transient Reactor Conditions	ORNL	~ 100,000
3.	Properties of Fission Product Aerosols Produced by Overheated Reactor Fuels	ORNL	~ 100,000
4.	Characterization, Control and Simulation of Accident-Released Fission Products	ORNL	1-200,000
5.	Nuclear Safety Pilot Plant	ORNL	3-400,000
6.	Fission Product Release Studies	BNL	2-300,000
7.	Fission Product Deposition from Gas Streams on Surfaces	North Carolina State	< 100,000
8.	Deposition Characteristics of Accident-Released Fission-Product Iodine	BMI	< 100,000
9.	R&D on New Air-Cleaning Techniques and Testing Methods	Harvard Air Cleaning Lab	1-200,000
10.	Filter Evaluation Program	ORNL	1-200,000
11.	Off-Shore Siting	ORNL	~ 100,000
12.	Machine Program for Fission Product Release	Phillips Petroleum Co.	< 100,000
13.	Code for Transport and Deposition of Fission Products	Phillips Petroleum Co.	< 100,000

Table 2. Gas-Cooled Reactor Program.
Research Tasks Relating to Fission Product Behavior

No.	Title	Sponsor	\$ in FY 66
1.	Steam-Carbon Reaction and Fission Product Release and Transport Studies	ORNL	< 400,000
2.	In-Pile Studies of the Reactions of Fueled Graphite and the Behavior of Fission Products Under HTGR Accident Conditions	ORNL	~ 100,000
3.	HTGR Large-Scale Steam-Graphite Test	ORNL	1-200,000
4.	ORR Poolside Capsule Irradiations for HTGR Safety Program	ORNL	~ 100,000
5.	High Temperature Behavior of Gas-Borne Fission Products	ORNL	< 100,000

Table 3. Fast Sodium Reactor Program
Research Tasks Relating to Fission Product Behavior

No.	Title	Sponsor	\$ in FY 66
1.	Chemical Reactions (Fast Sodium-Cooled Systems)	ANL	3-400,000
2.	Fission Product Retention by Reactor Coolants	Atomics Int.	< 100,000
3.	Fission Product and Contamination Control	Atomics Int.	< 100,000
4.	Characterization of Aerosols Generated from Explosive Spray of Sodium	Atomics Int.	< 100,000
5.	Characterization of Energy and Aerosols Generated from Sodium Pool Fires	Atomics Int.	3-400,000

THE ORNL RESEARCH AND DEVELOPMENT PROGRAM

ORNL conducts about half of the Research and Development tasks listed in the first three tables. Since ORNL also has the second largest budgetary fund of all the Commission's Nuclear Safety Program Contractors, you might be interested in a breakdown of our activities. Table 4 shows how the program is organized, with the work being divided among four of the Laboratory's divisions. As you can see, the major portion of the work falls in two divisions, Reactor Chemistry Division under George Watson, and Reactor Division which is my responsibility.

All of the work in the Reactor Chemistry Division is relevant to this meeting; on the other hand much of the work in the other divisions is paper evaluations or R&D on other aspects of Nuclear Safety. Two of the most important activities in the Reactor Division's program are the Nuclear Safety Information Center (NSIC) and the Nuclear Safety Journal, both of which were discussed by J. P. Blakely in a paper² at this meeting yesterday. Three groups in the Reactor Division perform work of relevance to my topic this morning; these are the Nuclear Safety Pilot Plant, the Engineering Scale Test and the ORR Poolside Capsule Tests groups.

I would like first to describe some of the facilities which are employed in this work and then to review some of the recent results.³ There are, however, two important concepts which must first be appreciated in order to understand the program and how it hopes to attain its objective of defining fission product behavior adequately, so that the results may be confidently, and conservatively extrapolated to actual reactor systems. The first concept is the sequence of events common to fission product behavior in all nuclear incidents. These are shown schematically in Fig. 1 and from left to right include,

- 1) the release of fission products from a fuel element
- 2) the transport of fission products from the point of release to the containment vessel
- 3) the behavior of fission products in the containment vessel, and
- 4) the removal of fission products in gas cleaning systems.

Table 4. ORNL Nuclear Safety Program Organization

NUCLEAR SAFETY PROGRAM: WM. B. COTTRELL, COORDINATOR

Figure 1 Life History of Fission Products During a Reactor Accident

Each of the experimental facilities that I will describe, routinely provides data on one or more of these processes which follows a major accident.

The other important concept relates to the geometric size of the various facilities. The results of the small scale experiments must be extrapolated to large sizes, in order to provide useful information for reactor safety analyses. The extrapolation of small scale experiments must be extensively tested to ascertain that the predominant mechanisms do not change with the size of the experiment. Accordingly, you will note in the discussion that follows, that some overlapping is involved in work performed in the various facilities. This is not accidental. It is done on purpose to test the validity of extrapolations with scaleup.

Such scaleup is necessary because the large experiments which are planned in the U.S. Nuclear Safety Program - such as the LOFT Experiment - will be exceedingly complex, costly, and necessarily few in number. The interpretation of their results will, therefore, depend heavily on a wealth of information obtained from many smaller scale experiments.

Figure 2 lists the major facilities and/or programs in the overall ORNL Nuclear Safety Research and Development effort. We are concerned here with primarily the items listed under capital 'A'. The in-pile experiments involve the greatest geometric extrapolation because of their small size, but introduce the least uncertainty regarding the adequacy of the heat source simulation. The three out-of-pile facilities include containment volumes ranging in size from $\sim 50 \text{ ft}^3$ for the CMF, 140 ft^3 for the CRI to 1350 ft^3 for the NSPP. In all instances the fission product release from the UO_2 fuel must be effected in a non-nuclear furnace. The first two facilities employ induction heating and the last a plasma torch.

Figure 3 shows the in-pile reactor furnace for simulating loss-of-coolant fuel destruction experiments. This 2-1/2" diam capsule contains a miniature fuel element, and is inserted into one of the fuel holes in the Oak Ridge Research Reactor where the fuel element melts under its own fission heat.

During the experiment, gas (which may be inert, or may be air, or steam, etc.) passes over the fuel element and helps carry away some of the

ORNL-DWG 66-2118

- I. Present Facilities and Programs of the ORNL Nuclear Safety Project
 - A. Release Transport and Deposition of Fission Products
 - 1. Out-of-Pile
 - (a) Containment Mockup Facility (CMF)
 - (b) Containment Research Installation (CRI)
 - (c) Nuclear Safety Pilot Plant (NSPP)
 - 2. In-Pile
 - (a) TREAT Experiments
 - (b) Loss-of-Coolant Fuel Destruction Experiments
 - B. Characterization and Simulation of Aerosols
 - C. Identification of Iodine Compounds
 - D. Iodine Removal From Gas Streams

Fig. 2 Facilities and Projects in the ORNL Nuclear Safety Program

Fig. 3 In-Pile Capsule

released fission product aerosol, which also contains some vaporized fuel and stainless steel or Zircaloy fuel cladding.

The aerosol is characterized as it goes through the small exit tube which doubles as a diffusion tube and the aerosol then goes to a filter compact. Figure 4 shows a schematic of the In-Pile Experimental Assembly. The reactor furnace that was shown in Fig. 3 fits right in its place in a position in the ORR reactor here shown at the bottom of the figure. In this assembly we can see the furnace in the reactor core, the filter, and the hydraulic mechanism that inserts it into the proper flux in the reactor.

These and other in-pile destruction studies are highly successful, and they have included experiments to study effect of atmosphere and condensing steam, effect of burnup, of gas flow rate, of nature of fuel, and of method of fuel destruction. In addition, data has been obtained on fission product fractionation, and freezing point depression of UO_2 by oxidized stainless steel, which causes depressions as large as 1000°C in the melting point of UO_2 .

The TREAT capsule facility is conceptually quite similar, although in detail significantly different. The purpose of the TREAT experiments is, of course, to simulate fission product release from accidents due to nuclear transients. The components of the experimental facility simulate on small scale a fuel element, the pressure vessel, and the containment system.

The TREAT experiments have been performed in atmospheres of argon, air-steam, steam at high pressure, and under water. The tests have employed UO_2 with stainless steel and Zircaloy, and in various tests have included partial as well as complete melts. For comparable melts, the fission product release in the TREAT experiments has always been significantly less than the ORR fuel melts, primarily due to the time-temperature characteristic of the heating transient.

Going on to the out-of-pile programs, Fig. 5 shows the diagram of the Containment Mockup Facility. It is built in a hot cell in order to handle large quantities of activity release when previously irradiated fuel is melted. The facility has a furnace to melt the fuel, and generates an aerosol cloud, which passes directly into a 180-liter containment vessel. In

Figure 4 Fuel Meltdown Experiment Assembly in Reactor

Figure 5 Schematic of Containment Mockup Facility

the containment vessel, the aerosol is aged for a pre-determined period and during the aging period samples are taken of the containment atmosphere to determine how the concentration of aerosols changes with time. After the aerosol is aged, it is displaced by argon introduced at the bottom. As the aerosol is displaced either by pressure release or by argon, it is passed through sampling and/or filtering devices depending upon the experiment. Figure 6 shows typical results of the containment atmosphere as function of time, as a simulated high burnup (10,000 Mwd/ton) Zircaloy-clad UO_2 fuel was melted in steam-air atmosphere. It is noted that even after 5 hours iodine and cesium still are present in the tank atmosphere to the extent of some 2 percent of the initial release.

Figure 7 shows a schematic of the Containment Research Installation (CRI) which has very recently been placed into operation. This installation has (1) a primary vessel of 120-liter capacity that can be pressurized and heated to 800°C ; (2) it has a containment vessel of 4000-liter capacity that can be pressurized and supplied with different surfaces - i.e., painted or concrete surfaces; (3) it has an aerosol generator capable of handling 100 grams of fuel; (4) it has capabilities for simulation of top or bottom breaks in the primary reactor piping. This facility has just recently been completed and no experimental data are available as yet. This facility was designed with a number of experimental capabilities including (1) release of fission products from the fuel, (2) retention in primary systems, (3) transport to the containment vessel, (4) behavior of fission products in the containment vessel, as well as, (5) recycle filter systems, (6) pressure suppression devices, and (7) spray and dousing techniques which may be simulated in the containment vessel.

Figure 8 shows a flow diagram of our largest facility - the Nuclear Safety Pilot Plant. The Nuclear Safety Pilot Plant is used to study the behavior of fission product aerosols in a containment vessel of substantial size (about 40,000 liters) to determine whether a change in scale has had a significant effect in scaleup extrapolation. It can handle fuel elements that are 1-in. in diam $\times$ 6-in. long and 250 grams in weight and uses a plasma jet as a source of heat. It employs many advanced techniques in sequential sampling and has all of the experimental capabilities of the CRI, except that for changing the inner surface of the vessel.

Figure 6 Percent of Total Inventory Airborne

Figure 7. Elevation of Containment Research Installation

ORNL-DWG 64-8767

Figure 8. Schematic of Nuclear Safety Pilot Plant

I shall now summarize some of the fission product release results and other data which have been obtained in these facilities.⁴ However, in order to compare the data, one must first clearly specify what the fission products are released from, and it has been found convenient to distinguish between those fission products which are released from the fuel itself, and those which are released from a zone surrounding the fuel which has a nominal temperature around 1000°C. In general, while large quantities of fission products are released from the fuel, much smaller amounts are released from this high temperature zone. For example, 99% of the volatile iodine, tellurium, and cesium and about 70% of the non-volatile strontium, barium, zirconium, and cerium are released from the fuel zone when the fuel is melted, but only about 85% of the volatile materials and generally less than 1% of the non-volatile materials are released from the high temperature zone. This trapping of fission products by high temperature surfaces is very significant. The physical structure and limitations of the experimental apparatus used for some types of experiments prevents precise definition of such release zones. However, these release values tend to agree remarkably well with the values for the release from the high temperature zone.

Table 5 shows the fission product release from UO_2 melted in an inert atmosphere. In general, I will classify the fission products into three groups: the volatile materials iodine, tellurium, and cesium, a changeable group consisting only of ruthenium, and the non-volatile materials strontium, barium, zirconium, and cerium. The top row of numbers will be shown in some subsequent figures for comparison purposes. For any figure showing results from the loss-of-coolant experiments in the ORR, or from the transient experiments in TREAT, the values given are release percentages from the high temperature zone, while the values from other types of experiments are release from less well defined zones. The release values, however, are seen to agree remarkably well. The high value for ruthenium from the out-of-pile experiment using an arc-image furnace is due to the presence of oxidizing impurities in the helium. The somewhat low values for the volatile materials released from the out-of-pile experiment using the central rod heater are due to the incomplete melting of the fuel. These values are comparable to those from loss-of-coolant experiments in which less than 20% melting occurred.

Table 5 Fission Product Release from UO_2
Melted in a Helium Atmosphere

Type of Experiment	Percent of Fission Product Released					
	I	Te	Cs	Ru	Sr-Ba	Zr-Ce
Loss-of-coolant experiments in ORR, 100% melted, stainless steel clad	90	76	78	3	1	0.3
Out-of-pile experiment using an arc image furnace and a BeO holder (impure helium)	90	92	91	61	3	2
Out-of-pile experiment using induction furnace and a tungsten crucible	95	92	72	0.3	2	0.1
Out-of-pile experiment using tungsten center rod heater, 56% melted, stainless steel clad	52	73	46	0.5	2	0.1

Table 6 shows fission product release from UO_2 melted in an oxidizing atmosphere. Note the first line for comparison purposes. The oxidizing or reducing capabilities of atmospheres such as steam-air mixtures depends on the relative amount of each component in the mixture since stainless steel reacts with steam above 1500°C to form hydrogen. Thus, the operating conditions of each experiment were qualitatively evaluated in order to properly classify the atmosphere. Except for the last two types of experiments, the data appear to agree fairly well. The CMF experiments are evidently difficult to classify. The releases of iodine, tellurium, and cesium seem to indicate that the experiments are classified correctly, but the ruthenium value indicates reducing conditions. The low values for the TREAT experiments are probably due to the extremely short transient melt time.

Table 7 shows release from UO_2 melted in a reducing atmosphere. Again note the first line for comparison. The values generally agree fairly well although some NSPP values are different, most probably due to incomplete melting.

From these and other data we can determine the amounts of the fission products that are retained in the fuel and in the high temperature zone for different environments. The release of fission products has also been shown to be not greatly affected by burnup or by sweep gas velocity, even though it is significantly affected by the composition of the atmosphere and by the effects of atmosphere on the cladding. It is also significant that fission product release data obtained from different types of experiments are in reasonably good agreement. Among the papers to be presented in this session are several which will present more detailed and more current data of this type.

Table 6 Fission Product Release from UO_2
Melted in an Oxidizing Atmosphere

Type of Experiment	Percent of Fission Product Released					
	I	Te	Cs	Ru	Sr-Ba	Zr-Ce
Loss-of-coolant experiments in ORR, 100% melted stainless steel clad UO_2 in helium	90	76	78	3	1	0.3
Loss-of-coolant experiments in ORR, 100% melted stainless steel clad UO_2 in moist air	97	72	68	15	1	0.4
Out-of-pile experiments, UO_2 melted in air using an arc image furnace	97	86	65	83	0.7	2
NSPP experiment, stainless steel clad UO_2 completely melted in an air-helium atmosphere using plasma torch	97		44	27	0.05	0.2
CMF experiment; stainless steel clad UO_2 melted in air or steam- air atmosphere, using an induction furnace	85	28	38	0.2	0.03	
TREAT experiment, stainless steel clad UO_2 melted in an air- steam atmosphere	5	0.8	3	0.7	3	0.5

Table 7 Fission Product Release from UO_2
Melted in a Reducing Atmosphere

Type of Experiment	Percent of Fission Product Released					
	I	Te	Cs	Ru	Sr-Ba	Zr-Ce
Loss-of-coolant experiments in ORR, 100% melted stainless steel clad UO_2 in helium	90	76	78	3	1	0.3
Loss-of-coolant experiments in ORR, 100% melted stainless steel clad UO_2 in steam-hydrogen-helium	66	87	47	0.3	2	0.2
Loss-of-coolant experiments in ORR, 100% melted stainless steel clad UO_2 in steam-air	67	92	42	0.8	0.8	0.1
NSPP experiment: stainless steel clad UO_2 melted in a steam- hydrogen-helium atmosphere using plasma torch	53	31	43	9	0.5	0.3

FISSION PRODUCT RELEASE IN ACTUAL INCIDENTS

It is with some reluctance that I review this work on the release of fission products before a group that may not have a pretty keen perspective into the technical and administrative problems of nuclear safety technology. Without such a perspective, some people may be tempted to jump to the conclusion that the large release fractions which have been obtained in some experiments can also be expected to provide the source term in reactor accidents. I do not believe that to be the case. What we have shown in the preceding experiments is that in an accident in which core melting occurs, fission products from molten fuel elements may escape from the fuel and the high temperature core to the extent indicated. However, even if no remedial action is taken, not only is it unlikely that the entire core will be molten, but also after leaving the high temperature core, the fission products are exposed first to the low temperature (350°C) primary system and then to the "ambient" temperature containment vessel. The various fission product removal processes - deposition, condensation, agglomeration, etc. - which occur in these regions, are known to effect further substantial reductions in the amounts of the non-gaseous fission products which would be ultimately available for release in the event of such an accident. Several of the subsequent papers in this session will discuss experiments in which the effects of these removal processes are examined.

In addition to these experiments simulating the behavior of accident-released fission products, an examination of fission product behavior in actual reactor accidents is very reassuring; first because there have been so few reactor incidents and secondly, because the fractional fission product release in these incidents has been so small.

The information shown in Table 8 was assembled⁵ in an effort to draw together widely scattered data on fission-product behavior in actual reactor accidents. This list is not intended to include all reactor accidents, but rather only those in which any significant fission product release occurred. However, examination of this data shows that rather scanty data were available for some of the incidents included. Most of the reports on reactor incidents contain much more of the details of circumstances that

Table 8 Fission Products Released in Major Reactor Incidents

Reactor	Type	Date	Incident	Fission Product (curies)
1. X-10 Pile	GCR	1947-1948	Cladding Failures	Not reported
2. NRX	D ₂ O	12-52	Undercooling	10,000
3. NRX	D ₂ O	12-55	Waterlogging	Not reported
4. G-1	GCR	10-56	Undercooling	20 to 50
5. Windscale	GCR	10-57	Wigner Energy	30,000 I ¹³¹ , 12,000 Te ¹³² , 600 Cs ¹³⁷
6. EL-3	D ₂ O	4-58	Undercooling	100 Xe ¹³³ , others not reported
7. NRU	D ₂ O	5-58	Waterlogging	Not reported
8. OMRE	Organic	10-58	Undercooling	1000
9. SRE	Sodium	7-59	Cladding Failures	0.3%
10. WTR	PWR	4-60	Undercooling	5000
11. SL-1	BWR	1-61	Excursion	84 I ¹³¹ , others not reported
12. ETR	Water	12-61	Undercooling	~ 50
13. MTR	Water	11-62	Undercooling	Not reported
14. ORR	Water	7-63	Undercooling	< 3000
15. BONUS	BWR	11-64	Undercooling	Small
16. PRTR	PWR	9-65	Rod Swelling	Not reported

led to the accident than of its consequences. The dispersal of fission products in some incidents was of such a nature that only qualitative statements concerning fission-product behavior were possible, but, even in some of these cases, the qualitative information reported leaves much to be desired. It seems that, at most reactor installations that had reportable incidents, accident-released fission products were regarded as an annoyance, rather than as a source of information that might be useful to reactor designers and to operators of other reactors of the same type.

However, several conclusions seem to be warranted by the data in Table 8. The first is that every one of the accidents listed (the BORAX-IV incident, as noted in the table, cannot be considered an accident) involved fuel melting or burning or both. This observation seems to justify the large amount of effort currently being applied to the problem of assessing the hazard of loss-of-coolant accidents. It is also worthy to note that the fuel in most of these accidents was either metallic uranium or an alloy of uranium and that the cladding material in most cases was aluminum, a very low-melting material. Only two cases involved reactors employing high-melting fuel materials (e.g., UO_2 , m.p. 2850°C) clad with high-melting materials such as zirconium (m.p. $\sim 1900^\circ\text{C}$) or stainless steel (m.p. $\sim 1500^\circ\text{C}$), and in these the release was trivial. All this, despite the extensive numbers of megawatt-days of power, have been generated by reactors employing the oxide fuel (Shippingport, Dresden, Yankee, etc.).

It is interesting to find that evidence of a metal-water reaction was noted in only four of the incidents involving water-cooled reactors, NRX (1952), EL-3, NRU, and SL-1. In each case it appears that reaction of uranium or aluminum with water occurred, but not at a catastrophic rate, such as had been feared might occur under such circumstances. It seems unlikely that the chemical reaction contributed a significant fraction of the total energy involved in the first three incidents and may have been as much as 20% in the last.

Except for the Windscale incident, comparatively small quantities of released fission products other than the rare gases, escaped from the confines of the reactor buildings, although they were not designed primarily for containment. None of the incidents included in the table, however, approached the seriousness of the maximum credible accident.

As reassuring as such considerations may be regarding our ability to design, build and operate nuclear reactors safely, the safety record which has been compiled by the AEC and its contractors is even more so. Unfortunately, comparable statistics do not exist for commercial nuclear power facilities, but there is every reason to believe that their record is at least as good because it does not involve such extensive experimental programs as does the AEC operations. The data I will summarize briefly was prepared by the USAEC's Division of Operational Safety⁶ and covers the 22 years from 1943 to 1964.

Figure 9 shows the deaths and death rates in atomic energy installations and compares the rates with those for the overall employment force in the whole United States. Note the pleasing declining trend of both, and note that the average value for the AEC is 12.0 per 100,000 versus 25.7 for industry as a whole. Figure 10 shows the causes of the fatalities. Of the 251 deaths, 81 were caused by falls or falling objects, 62 by motor vehicles, 38 by electric shock, 15 by burns, and 3 by radiation. Note that this figure does not include the three fatalities at SL-1 as radiation fatalities since the immediate cause of the death of the three men in the SL-1 accident was explosion and missiles, but the radiation levels were sufficiently high to have caused their death in a short time. Figure 11 shows that the severity rates for all accidents of the AEC and its contractors is again very substantially less than that of the country as a whole. Figure 12 shows the total injuries over a 22-year period and a comparison with the very low number caused by radiation. Using the AEC figure of 36, this gives 0.47%. If the three SL-1 victims are added as radiation injuries, the injury percentage becomes 0.51. Note there has been only one fatality due to radiation in a privately-owned, licensed nuclear facility. Figure 13 indicates the time lost by AEC and its contractors by accidents. Note that not only is the time lost for all accidents very low but those due to radiation are a small fraction of the total (20,124 days out of 388,803,937 days, or 0.0052%). Again, adding 18,000 days more for the three SL-1 victims only makes the lost days from radiation 0.01% of the total man days.

ORNL-DWG 66-9063

Figure 9

Figure 10

ORNL-DWG 66-9065

Figure 11

RADIATION INJURIES 1943-1964

AEC OPERATION CONTRACTORS

Figure 12

● **TIME LOST from RADIATION INJURIES 1943-64**
AEC OPERATION CONTRACTORS

Figure 13

In the immediately preceding discussion we talked about fatalities and their causes, injuries and their severity, and the amount of time lost as a result of them. It has been shown by a period of 22 years of experience that radiation can be handled with injuries to the workers at a much lower rate and of far less severity than those due to falls, falling objects, electric shock, burns, and motor vehicles. It is hard to escape the conclusion that if we want to improve the safety of the radiation workers, the place to put the effort is on these other causes while at the same time maintaining and improving the present system of radiation monitoring and control.

At this point we should look at the radiation exposure of the public and the environment from reactor accidents. However, except for information on natural background radiation and some data on fall-out from weapons testing, I am unaware of any meaningful data. From the characteristics of radiation and radioactive material, it is certain that the exposure of the public is at least one or two orders of magnitude lower than the exposures to radiation workers. In the years to come perhaps there will be data, but probably not. This is because, it is to be expected that the present care in handling radiation and radioactive material will be continued and also because any radiation levels in the environment are likely to be so low, that they cannot be measured with any precision.

DISCUSSION AND CONCLUSIONS

In my talk I first summarized briefly some of the typical results that have been obtained to date from experiments on the behavior of accident released fission products. Following that, I have attempted to put the reactor accident potential in perspective in terms of the historic data on radiation injury to reactor operators and the public at large. Within our limited experience, the data from accident-simulating experiments as well as from all known reactor incidents is exceptionally good. The limited experience plus the difficulty of proving the negative argument -- that a severe accident cannot occur -- were the basis for the initiation of the research and development program on the behavior of released

fission products. However, concurrent with the general acceptance of the principle that consequence-limiting engineered safeguards are more demonstrable and therefore more acceptable than accident prevention or even accident-limiting safeguards, the objective of the R&D program has shifted from fission product release to fission product transport and removal.

As far as reactor accident analysis is concerned, the accuracy of fission product release estimates exceeds both the accuracy with which the core geometry and temperature distribution can be predicted as a function of time, as well as the present understanding of fission-product transport phenomena. Thus recent efforts in the field of fission-product behavior studies have been directed toward a better understanding of transport phenomena, both natural and induced. This, in turn, has necessitated the determination of the physical and chemical states of the released fission products (characterization) in order to understand the transport phenomena. Laboratory tests have already identified various forms of different fission-product elements, particularly iodine, and have indicated, if incompletely, their behavior under a variety of conditions simulating those which might exist in a reactor accident.

The behavior of released fission products is, however, expected to be a function of the geometry and environment (including materials and their surface conditions) in the containment system, as well as the nature of the released fission products themselves. In order to verify that the behavior of released fission products in actual containment systems may be extrapolated from existing laboratory-scale tests, three facilities, each permitting a closer approximate to a full-size reactor meltdown, have been, or are being built. These facilities are the Nuclear Safety Pilot Plant (NSPP) at ORNL, the Containment Systems Experiment (CSE) at Hanford, and the Loss-of-Flow Test (LOFT) at NRTS. Of these three facilities, only the NSPP is in operation and will be discussed in a later paper. If the fission product transport behavior in these facilities substantiates that which may be extrapolated and/or inferred from the laboratory tests now well underway, a much more realistic evaluation of the consequence of reactor accidents will become available. The experimental program may be expected to demonstrate that the fission-product inventory available for

leakage out of a containment system may be orders of magnitude less than that which is currently presumed in accident analyses for license applications, and this would be consistent with interpolations from the limited data now available from actual reactor accidents. Such conclusions would have a profound effect on reactor siting criteria and the requirements of engineered safeguards.

REFERENCES

1. J. R. Buchanan and N. F. Cross, Current Nuclear Safety Research and Development Projects, USAEC Report ORNL-NSIC-10, Oak Ridge National Laboratory, June 1966.
2. J. P. Blakely, How to Find Answers to Questions on Nuclear Safety, paper presented at the 152nd National Meeting of the American Chemical Society, New York City, September 12-16, 1966.
3. Readers interested in more details on the ORNL Nuclear Safety Program should read the semiannual progress reports (now being changed to an annual report). The last report in this series is listed below; prior reports are listed in each report. Nuclear Safety Program Annual Progress Report for Period Ending December 31, 1966, USAEC Report ORNL-4071, Oak Ridge National Laboratory, March 1967.
4. Most of the data here was taken from the following paper but is also available in reference 3. B. F. Roberts, Evaluation of Fission Product Release Data, paper presented at ORNL Nuclear Safety Program Annual Information Meeting, Oak Ridge, Tennessee, Feb. 13-15, 1967.
5. C. J. Barton and G. W. Parker, Behavior of Released Fission Products in Actual Reactor Incidents, Nuclear Safety 5(4): 389 (Summer 1964).
6. Division of Operational Safety, Operational Accidents and Radiation Exposure Experience Within the United States Atomic Energy Commission, April 1965.

INTERNAL DISTRIBUTION

1. R. E. Adams	34-38. Nuclear Safety Information Center
2. S. E. Beall	39. L. G. Overholser
3. J. P. Blakely	40. G. W. Parker
4. R. E. Blanco	41. L. F. Parsly
5. W. E. Browning, Jr.	42. A. M. Perry
6. C. A. Burchsted	43. M. W. Rosenthal
7-16. Wm. B. Cottrell	44. A. W. Savolainen
17. F. L. Culler, Jr.	45. L. B. Shappert
18. M. H. Fontana	46. O. Sisman
19. A. P. Fraas	47. M. J. Skinner
20. P. N. Haubenreich	48. B. A. Soldano
21. P. R. Kasten	49. I. Spiewak
22. S. I. Kaplan	50. D. B. Trauger
23-24. G. W. Keilholtz	51. G. M. Watson
25. M. I. Lundin	52. G. D. Whitman
26. R. N. Lyon	53-54. Central Research Library (CRL)
27-28. R. E. MacPherson	55-56. Y-12 Document Reference Section (DRS)
29. A. P. Malinauskas	57-59. Laboratory Records Department (LRD)
30. H. C. McCurdy	60. Laboratory Records Department-Record Copy (LRD-RC)
31. J. W. Michel	
32. A. J. Miller	
33. C. E. Miller, Jr.	

EXTERNAL DISTRIBUTION

61. William W. Lowe, Suite 1104, 1730 M. Street, NW, Washington, D. C. 20036
62. Peter F. Santoro, Combustion Engineering, Inc. Box 500, Windsor, Connecticut 06095
63. P. Sinder, U.S. Atomic Energy Commission Depository Library, 7 Woodland Avenue, Larchmont, N.Y. 10538
64. J. M. Skarpelos, Vallecitos Atomic Laboratory, General Electric Company, Box 846, Pleasanton, California
65. S. R. Smith, Savannah River Laboratory, Aiken, S.C. 29802
- 66-80. Division of Technical Information Extension (DTIE)
81. Research and Development Division, ORO
- 82-83. Reactor Division, ORO