

**Reactor Safety Research**  
**Summary of Activities and Results – FY84**  
**for**  
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## 2. FISSION-PRODUCT SOURCE TERM

### 2.1 HIGH-TEMPERATURE FISSION-PRODUCT CHEMISTRY

(D. A. Powers, 6422; R. M. Elrick, 6422; R. A. Sallach, 1846)

Release of radionuclides from the reactor fuel during a severe accident is but the first phenomenon that must occur to create an inventory of radionuclides available for release from the plant. Once released from the fuel, the radionuclides must travel to a point where they can escape the plant before these radionuclides have any safety consequences. Two processes can limit the efficiency of radionuclide transport within the reactor coolant system. Fission-products can react and bind to cool surfaces of structural materials within the reactor coolant system. Aerosol processes can cause radionuclides to deposit or sediment on structures within the coolant system and eliminate these radionuclides from the inventory of releasable radioactivity.

The objective of the High-Temperature Fission-Product Chemistry program is to experimentally characterize the chemical interactions of radionuclides that should affect the efficiency of radionuclide transport in reactor coolant systems. Most of the work in this program has been focused on the reactions of volatile fission products such as Cs, I, and Te with structural materials. Results obtained in the quantitative characterization of these chemical processes are used in reactor accident source term analyses such as that sponsored by the NRC's Accident Source Term Project Office. The tellurium retention on structures predicted in these analyses (the analyses are commonly called BMI-2104) is based on deposition velocities obtained in the High-Temperature Fission-Product Chemistry program. Data obtained in this program on CsOH vaporization have also had an important use in the source term analyses sponsored by the IDCOR group.

Recently, it has been recognized that radionuclides deposited on structures will heat these structures because of radionuclide decay. Heating of structures may be sufficient to cause deposited radionuclides to revaporize and again be available for release from the plant. Results obtained in the High-Temperature Fission-Product Chemistry program have a significant bearing on such predictions. Experimental studies in this program have shown that deposited radionuclides undergo significant chemical transformations. CsOH is converted to cesium silicate, cesium phosphate, and the like. Tellurium is converted to metal tellurides. These chemical transformations alter the volatility of the radionuclides-frequently reducing the volatility. Analyses of revaporization phenomena in severe accidents have been, to date, scoping in nature and have not

taken into account chemical transformations of deposited radionuclides. Chemical transformations may be more important than gas flow for revaporization. More realistic analyses of revaporization will require data such as that generated by the High-Temperature Fission-Product Chemistry program.

The activities in the High-Temperature Fission-Product Chemistry program during the year were focused in the following areas:

1. Definition of the chemical form of cesium deposited on stainless steel;
2. Interactions of cesium hydroxide and cesium iodide with boron carbide in steam; and
3. Effects of simultaneous steel oxidation on the deposition of tellurium.

2.1.1 Definition of the Chemical Form of Cesium Deposited on Stainless Steel

Very detailed studies of the microstructure and chemical composition of stainless steel exposed to cesium hydroxide vapors in steam and hydrogen were undertaken this year. The objective of these studies was to ascertain the chemical nature of the so-called "water insoluble form" of deposited CsOH. Cesium hydroxide has been shown in this program to deposit on stainless steel in two forms. One form is readily removed by (1) temperatures in excess of about 800°C, and (2) water leaching the specimen. The other form of deposited cesium hydroxide is not water soluble. This water insoluble form persists on stainless steel coupons at temperatures up to at least 1000°C. This is the form of cesium found on lead screws taken from the reactor at Three Mile Island Unit 2.

Quantitative analyses of the elemental distributions within oxide coating formed on stainless steel in steam show that cesium is present as cesium silicate ( $\text{Cs}_2\text{Si}_4\text{O}_9$ ). Silica is formed in the oxide by steam oxidation of the Si impurity in stainless steel. The presence of cesium silicate rather than cesium chromate may be surprising to some familiar with the literature of fuel/clad interactions for fast reactors. Cesium chromate is not stable in the steam-hydrogen atmospheres of interest for radionuclide transport in the reactor coolant system during LWR accidents.

Demonstration of the chemical form of deposited cesium is of major significance in reactor accident source term analyses. Quite clearly, cesium bound in the steel as cesium silicate is not susceptible to mechanical resuspension as a result of vessel depressurization or in-vessel

steam explosions. The susceptibility of cesium silicate to revaporization as a result of temperature excursions of the steel may also be limited. Analyses to date of cesium revaporization have presumed that the cesium remains in the form of cesium hydroxide, which is quite volatile. Results of the work in the High-Temperature Fission-Product Chemistry program show deposited cesium is transformed chemically to a much less volatile form. At worst, much higher temperatures will be required to revaporize cesium from cesium silicate than from cesium hydroxide. The timing and even the magnitude of cesium revaporization then should be quite different than has been supposed up to now.

Identification of cesium silicate also calls into question the mathematical description of fission-product deposition found in accident analysis codes such as TRAPMELT. These codes hypothesize that the rate of deposition is proportional to the vapor phase concentration. Other limiting factors exist. For one, the availability of silica to react to form cesium silicate imposes a limit on the extent of cesium deposition. Further, the rate of deposition must approach zero when the vapor concentration of cesium approaches the cesium vapor concentration in equilibrium with cesium silicate.

The importance of having proper chemical characterization of deposited radionuclides for revaporization analyses was graphically demonstrated during the year. Industry analyses of revaporization using literature data for the vapor pressure of cesium hydroxide and more recent measurements obtained in this program have been done. Results of the analyses show that the lower vapor pressures measured in the High-Temperature Fission-Product Chemistry program reduce the importance of revaporization.

Quite clearly, the detailed studies of the chemical form of deposited cesium conducted in the High-Temperature Fission-Product Chemistry program have had remarkable effects on the quantitative analysis of radionuclide behavior in severe accidents.

#### 2.1.2 Interactions of CsOH and CsI with B<sub>4</sub>C in Steam

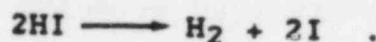
A "conventional wisdom" is growing that iodine released from degrading fuel will quickly become an iodide and be subject to aerosol physical processes. Such aerosol processes will mitigate the consequences of iodine release from the fuel. The conviction that iodine will be in the form of CsI after release from the fuel is based, to a large extent, on results of simplistic thermochemical analyses of the Cs-I-O-H system. Such simplistic calculations fail to take into account the wide diversity of material encountered when Cs and I are released into the atmosphere surrounding a degrading core.



Work in the High-Temperature Fission-Product Chemistry program has demonstrated several instances where the preferred form of iodine and cesium is not CsI, as held by the "conventional wisdom". One of these instances is particularly important for Boiling Water Reactors (BWRs). CsI will react with the products of steam oxidation of B<sub>4</sub>C control blades according to a formal stoichiometry that is approximately:



The gaseous species HI is susceptible to the equilibrium



Detailed thermochemical analyses of the B-Cs-I-O-H system were reported this year by investigators in the High-Temperature Fission-Product Chemistry program. These analyses show that the reaction requires some dilution of the activity of either CsBO<sub>2</sub> or HI to proceed significantly in the direction of CsI dissociation. One easy way to reduce the activity of CsBO<sub>2</sub> is to simply dissolve it in another oxide such as condensed phase B<sub>2</sub>O<sub>3</sub> that forms on B<sub>4</sub>C exposed to steam.

Results of the thermochemical calculations show that cesium activity, especially if it reacts with boron-bearing species, will be reduced relative to that of CsOH. This is yet another reason revaporization analyses hypothesizing the chemical form of cesium to be CsOH would be in error. The behavior of I released by reactions of CsI is not yet well understood. Clearly, it passed through the reaction system in experiments to date. It might not be so mobile in the complex chemical mixture flowing out of the reactor in a severe accident.

Another known cause of CsI dissociation is the photolysis of CsI in a radiation field. Cesium is readily ionized by gamma rays. Cesium iodide, when ionized, dissociates to yield Cs<sup>+</sup> and atomic iodine. In the environment of a reactor accident, an intense flux of gamma irradiation would be present and a quasi-steady state concentration of Cs<sup>+</sup> and consequently I would be expected. Thermochemical calculations to date have failed to take into account the photolysis processes. Because the formation and destruction of ions is a dynamic process, it cannot be treated within a strictly thermodynamic formalism. Rather, a set of unknown kinetic parameters are required.

To evaluate the importance of gamma photolysis on the chemical form of iodine in a severe reactor accident, a series of tests are being planned. The test series will consist of identical tests in laboratory environments and in an intense gamma irradiation field created by the decay of  $^{60}\text{Co}$ . The tests will evaluate the effects of irradiation on both the photolysis and the revaporization of  $\text{CsI}$ . The first laboratory runs for the test program have been conducted. Results of tests in the irradiation field could alter dramatically the conventional wisdom on the chemical form of iodine.

#### 2.1.3 Effects of Simultaneous Steel Oxidation on Tellurium Deposition

Tests of the simultaneous oxidation and tellurium deposition on stainless steel were conducted. Tellurium was found to be bound to nickel nodules in the oxide on the stainless steel rather than as a mixture of iron and nickel tellurides as had been observed previously. This observation again shows that fission-product deposition on surfaces in a reactor coolant system may not be strictly a function of vapor concentration. The availability of reactive surfaces, in this case nickel, also may be rate-controlling.

#### 2.1.4 Accident Source Term Project Office (ASTPO) Related Activities

An important aspect of the NRC's Source Term Reassessment effort has been the peer review and review by the American Physical Society of the accident analyses reported in the documents commonly known as BMI-2104. Investigators for the High-Temperature Fission-Product Chemistry program were diverted to preparing voluminous responses to hosts of technical inquiries made by members of the APS review committee. Detailed responses were made to the inquiries concerning the following topics:

1. Chemical form of radionuclides in fuel and in the fuel/clad gap;
2. Reactions of  $\text{CsOH}$  and  $\text{CsI}$  with  $\text{B}_4\text{C}$ ;
3. Solubility of fission products in high-temperature steam;
4. Chemical state of Ba and Sr during fuel/clad interactions;
5. Basis for CORSOR rate equations;

6. Effects of oxygen potential and pressure on fission-product release;
7. Revaporization of deposited radionuclides;
8. Effects of radiation and radiation-induced nitric acid formation on the chemical form of iodine; and
9. Resuspension of deposited aerosols in the reactor coolant system.

#### 2.1.5 Other Activities

Investigators in the High-Temperature Fission-Product Chemistry program are obligated to assist investigators involved with the PBF severe fuel damage experiments in interpretations of their chemical data. To this end:

1. Thermochemical analyses of the Cs-I-O-H system were conducted to determine the chemical form of radionuclide species during transit through the flow network at PBF.
2. Modifications were proposed to the CORSOR code to incorporate the effects of pressure and flow on the release of radionuclides as a possible explanation of differences between release results obtained out-of-pile at ORNL and in the PBF 1-1 tests.
3. A model of reverse reaction and flow stagnation was formulated to interpret results of the test PBF 1-3.

The models of fission-product release have a larger significance whether or not they are indeed applicable to the interpretation of complex, integral tests such as those done at PBF. These models incorporate physical and chemical processes not recognized in the calculations done for the NRC's source term reassessment, but expected to be operative in accidents. These processes will be most important for those accidents in which core degradation takes place in a pressurized vessel such as the TMLB' and TC sequences. Unfortunately, these accidents are often the risk-dominant accidents for many plants. Quite accurate source term estimates are most desirable for these accidents. The effects described by the models depress release rates. Thus, real releases may be much lower than anticipated in the BMI-2104 calculations. Aerosol processes may then be less efficient and retention of radionuclides within the reactor coolant system may be less important.

Also during the year investigators in the High-Temperature Fission-Product Chemistry program were called upon to review documents produced by IDCOR and to participate in meetings with IDCOR dealing with source term

concerns. Reviewing the documentation produced by IDCOR was, of course, an unprogrammed task that required a remarkable amount of time. Critiques of the IDCOR work were presented at meetings with IDCOR. Among the comments concerning the IDCOR work as it relates to research in the High-Temperature Fission-Product Chemistry program were:

1. Chemical transformation of CsI by reaction with borates ought to be considered in sequences for BWRs.
2. Chemical transformation of deposited species ought to be recognized in revaporization analyses.
3. Revaporization of Te ought to be considered.

Finally, investigators working on the High-Temperature Fission-Product Chemistry program participated in the preparation of a document commonly known as NUREG-1053. This document is intended to summarize the state of knowledge concerning source term phenomena, the data needs, and the modeling capabilities. Specific responsibilities were assumed by investigators in the program to summarize knowledge about fission-product release during core degradation. A draft of the chapter on this topic was produced. It is a rather thorough review containing in excess of 200 references dealing with both experiments and modeling. The review concludes with an importance ranking of information needs and an indication of where these needs will be met.

#### 2.1.6 Documentation

R. Sallach, Vapor Pressure of Liquid CsOH, SAND84-1693, Sandia National Laboratories, Albuquerque, NM, 1984.

R. A. Sallach, R. M. Elrick, A. L. Ouellette, and S. C. Douglas, Reactions Between Some Cesium-Iodine Compounds and the Reactor Materials 304 Stainless Steel, Inconel 600 and Silver, Volume I, Cesium Hydroxide Reactions, NUREG/CR-3197, SAND83-0395, Sandia National Laboratories, Albuquerque, NM, 1984.

R. A. Sallach, Calculated Vapor Compositions for the H<sub>2</sub>O+I+Cs+B System, SAND84-0662, Sandia National Laboratories, Albuquerque, NM, 1984.

R. A. Sallach, Chemical Aspects of CsI Interaction in Steam with 304 Stainless Steel and Inconel 600, SAND84-0749, Sandia National Laboratories, Albuquerque, NM, 1984.



R. A. Sallach and R. M. Elrick, "Chemical Reactions of CsOH, CsI and Te Vapors with Oxidizing Reactor Materials," Proceedings of the Topical Meeting on Fission-Product Behavior and Source Term Research, Snowbird, UT, July 15-19, 1984.

A. R. Taig, "Release and Retention Phenomena in Degraded LWR Cores," Proceedings of the Topical Meeting on Fission-Product Behavior and Source Term Research, Snowbird, UT, July 15-17, 1984.

A. R. Taig, "Interpretation of Experimental Results on the Interactions of Fission-Product Vapors with Reactor Materials," Proceedings of the Topical Meeting on Fission-Product Behavior and Source Term Research, Snowbird, UT, July 15-17, 1984.

R. A. Sallach, C. J. Greenholt, and A. R. Taig, Chemical Interaction of Tellurium Vapors with Reactor Materials, NUREG/CR-2921, SAND82-1145, Sandia National Laboratories, Albuquerque, NM, March 1984.

D. A. Powers, Release of Radionuclides During Core Degradation, Chapter 3, NUREG-1053.

#### 2.1.7 Meetings Attended

Severe Fuel Damage Meeting, October 29-November 2, 1983, Bethesda, MD.

11th LWR Safety Research Information Meeting, October 22-26, 1983, Gaithersburg, MD.

American Physical Society Review Meeting, January 15-18, 1984, Boston, MA.

American Physical Society Review Meeting, March 22-24, 1984, Albuquerque, NM.

Review of IDCOR Source Term Analyses, February 6-10, 1984, Hunt Valley, MD.

Review of IDCOR Source Term Analyses, May 13-15, 1984, Rockville, MD.

Severe Fuel Damage Meeting, April 10-11, 1984, Albuquerque, NM.

Review of IDCOR Analyses including Source Term Considerations, August 27-29, 1984, Rockville, MD.

Peer Review BMI-2104, January 25-27, 1984.

Peer Review BMI-2104, October 11-14, 1983.

Planning for NUREG-1053, June 4-5, 1984.

Planning for NUREG-1053, June 27-28, 1984.

## 2.2 QUANTITATIVE UNCERTAINTY ESTIMATE OF THE SOURCE TERM (QUEST) (D. A. Powers, 6422; P. K. Mast, 6425)

### 2.2.1 Objective of the Work

A key element of NRC research over the last two years has been the development of a new, mechanistic method for estimating radioactivity release from a nuclear power plant during a severe accident. The essence of this new method is the use of mechanistic computer codes to estimate the release and subsequent behavior of radionuclides in ways that explicitly recognize the peculiar features of plants and accidents in question. The objective of the new method is to provide more accurate and more realistic estimates of radioactivity release during severe reactor accidents than is possible using the prescriptive source term tabulations developed for the Reactor Safety Study.

As part of the development of the new method for source term estimation, a number of accident analyses were done and documented in a series of reports commonly known as BMI-2104. The computer codes used for these analyses were known, however, to be imperfect and input data for the codes were not known to great accuracy. A question then arises about the magnitude of the uncertainty that ought to be attached to source term estimates made using the new methods.

The objective of the Quantitative Uncertainty Estimate of the Source Term (QUEST) program was to develop an estimate of uncertainties that ought to be ascribed to mechanistic source term estimates using the currently available state-of-the-art computer models. Both the general magnitude of the uncertainties and the disposition of the uncertainty range about a point estimate of the source term was sought.

Two types of uncertainty were recognized in the QUEST study. The first type of uncertainty--the so-called  $\delta_c$  uncertainty--relates to the suite of computer codes utilized for the analyses of the accidents considered for the BMI-2104 documents. These computer codes represent the current state of the art. As the new method of source term estimation is accepted, these computer codes are what will be available for implementing the method. The codes rely, however, on a variety of input data--much of which is not known with precision. Different analysts may make, with good technical justification, different choices for the inputs to the suite of codes. The  $\delta_c$  uncertainty then provides an estimate