## RESEARCH INFORMATION LETTER (RIL) 1301

## FINDINGS AND USE OF RESULTS FROM PHÉBUS-FP TESTS TO VALIDATE THE NRC'S MELCOR SEVERE ACCIDENT CODE AND REVISED ACCIDENT SOURCE TERM (NUREG-1465)

### Background

The French Institut de Radioprotection et de Sûreté Nucléaire (IRSN) has conducted and is analyzing the suite of five Phébus Fission Product (Phébus-FP) integral experiments. This international cooperative research program is intended to develop experimental data for use in validating computer codes that are used to analyze severe reactor accidents. The U.S. Nuclear Regulatory Commission (NRC) is participating in the program to obtain the experimental data needed to validate the agency's MELCOR integral severe accident analysis code, as well as models used to quantify the long-term development of gaseous iodine in a pressurized water reactor (PWR) containment. The integral tests are being supplemented by four sets of separate effects tests in the Phébus International Source Term program (Phébus-ISTP) to help clarify the interpretation of the integral test findings. The four efforts include the BECARRE tests to investigate the interactions of boron carbide with the stainless steel cladding on control rods, the MOZART investigations of the interactions of air with zirconium alloy cladding used for fuel rods, the CHIP tests of fission product chemistry in the reactor coolant system under accident conditions, and the EPICUR tests of iodine behavior and interactions in a reactor containment. Furthermore, the Behaviour of Iodine (BIP) project has been initiated under the auspices of the Nuclear Energy Agency to conduct separate effects tests of iodine interactions with surfaces within the containment and in sumps. NRC is participating in both Phébus-ISTP and BIP.

The conduct of tests in the Phébus-FP experiment series is complete. The interpretation of the findings of all the tests are ongoing. It is possible, however, to address some of the more important findings of the test program and how the results are affecting the NRC's severe accident code. The findings also have implications for the NRC's revised NUREG entitled "Accident Source Terms for Light-Water Nuclear Power Plants" (NUREG-1465). Some of these findings and implications were discussed in the previous Phébus-FP Research Information Letters (RIL-0004 in 2000 and RIL-0702 in 2007). Additional insights have come from the separate-effects experimental programs (Phébus-ISTP and BIP) undertaken to support analysis and interpretation of the Phébus-FP integral tests.

As discussed in RIL-0702 the behavior of gaseous iodine in the containment model used for the Phébus-FP tests deviated some from expectations. As expected, the gaseous iodine concentration in the containment atmosphere dropped at an essentially exponential rate following a maximum during the period of the most rapid core degradation. This rapid decrease in gaseous iodine concentration was especially evident in the final test in which much of the iodine was released to the containment in gaseous form. The fall in gaseous iodine concentration did not continue, however, to a very low level. Instead, a low but steady concentration of gaseous iodine developed in the containment atmosphere and this steady state concentration persisted for days to the final termination of test observations. The steady-state concentration developed even when the model sump in the containment model was made deliberately basic and containment conditions were adjusted to promote steam condensation on the sump waters. These findings may have implications for Generic Safety Issue (GSI) 191, "Assessment of Debris Accumulation on PWR Sump Performance." On the basis of experiments reported in NUREG/CR-6913, "Chemical Effects Head-Loss Research in Support of Generic Safety Issue 191," interactions between chemicals used to control the pH of the sump waters and dissolved insulation materials dispersed into the sump can generate precipitates which exacerbate sump screen blockage. While eliminating sump buffer chemicals may be beneficial for sump strainer blockage concerns, chemical-effects testing has not been performed to address un-buffered sump pool environments. It was also observed in the integral effects tests that the chemical makeup of the steady state concentration of gaseous iodine varied between molecular iodine ($I_2$) and volatile organic iodide ($CH_3I$). The varying chemical makeup of the gaseous iodine may have implications on the possible use of filtered venting as a defense-in-depth safety strategy.

The EPICUR separate effects tests in the Phébus-ISTP program and the BIP program were undertaken to better understand the behavior of gaseous iodine observed in the Phébus-FP tests. The formulations of these programs were based on the working hypothesis which has largely been confirmed that the source of gaseous iodine in the Phébus-FP experiments was the iodine absorbed on epoxy painted surfaces within the containment model. These painted surfaces were on condensers incorporated into the model design to simulate painted surfaces of equipment and structures in reactor containments. Under irradiation the iodine absorbed in the paint re-evolves as both molecular iodine and volatile organic iodide. The gaseous species decompose due to both radiolysis and thermal reactions to form extremely fine aerosol particles. These particles redeposit on painted surfaces and the cycle begins again leading to a steady-state gaseous iodine concentration in the containment atmosphere. Predictions of the gaseous iodine concentrations hinge on the ability to scale results of the integral and separate effects tests to the geometries and conditions during reactor accidents.

## Regulatory Issue

Analyses of severe accident progression and fission product release have become especially important as the NRC modifies its reactor regulatory processes to become more risk-informed and performance-based. Integral experiments, such as the Phébus-FP tests, provide data that are especially useful for assessing the technical adequacy of the MELCOR computer code for use in Level 2 and Level 3 probabilistic risk assessments (PRAs) and in the support of risk-informed regulation.

In addition, the NRC's revised accident source term is expected to be used in offsite dose analyses for nuclear power plants to support operational flexibility, eliminate unnecessary conservatism, and assess the change in risk under Regulatory Guide 1.174, "An Approach for Using Probabilistic Risk Assessment in Risk-Informed Decisions on Plant-Specific Changes to the Licensing Basis." Some of the important features of the NRC's revised accident source term (NUREG-1465) have been confirmed by results of the Phébus-FP tests.

## Method

The Phébus-FP project consisted of five integral tests, all of which have been completed:

- The first test, FPT-0, was a "shake-down" test of the facility and used only trace-irradiated fuel. The fuel bundle did have a silver-indium-cadmium control rod similar to those used in PWRs. The purposes of the tests were to (1) verify the adequacy of the test procedures and instrumentation, and (2) provide information related to fuel degradation and fission product release, transport and behavior in a simplified model of a reactor coolant system and containment.

- The  second test, FPT-1, used intact fuel and cladding irradiated to 23 GWd/t. It was re-irradiated just prior to the test to build into the fuel detectable inventories of the shorter half-life radioisotopes. Again, a silver-indium-cadmium control rod was included in the test bundle.
- The third test, FPT-4, used fragment fuel irradiated to 32 GWd/t and zirconia shards in a debris bed similar to those formed during the accident at Three Mile Island. The purpose of the test was to simulate the later stages of core degradation in a severe reactor accident.
- The fourth test, FPT-2, was conducted with conditions for fuel degradation that involved less steam than in either test FPT-0 or test FPT-1. Otherwise, the test was similar to the FPT-1 test and did include a silver-indium-cadmium control rod. The intention of the test was to simulate steam-starved conditions similar to those that are expected to arise in some regions of a reactor core during an accident. Boric acid was injected into the test section to simulate the presence of borated water in PWRs.
- The final test, FPT-3, used irradiated fuel and a boron carbide control rod.

Post-test examinations of the all the tests save FPT-3 have been completed.

The EPICUR test facility consists of a flow system within a $^{60}Co$ irradiation facility. It allows solutions and solids contaminated with iodine to be irradiated under conditions of controlled temperature, relative humidity and flow velocity. The effluent gas can be sampled to distinguish the chemical form of any evolved iodine as either particulate iodides, molecular iodine, or volatile organic iodides. Tests have been performed to show that gaseous iodine absorbs on paint. Furthermore, the tests have shown that irradiated solutions of iodide ($I^-$) produce aqueous molecular iodine that absorbs on paint. The experiments also included irradiation of iodine-contaminated paint samples to determine how iodine can be released from paint. During irradiation, the iodine contamination re-evolves in the three distinguishable chemical forms. Typical tests last for 8 hours but some have involved irradiation for over 30 hours. Evolution rates vary with time and cease when irradiation stops.  Most of the tests in the EPICUR project used a Ripolin epoxy paint commonly found in French reactors.

The primary BIP test facility allows accurate measurement of iodine absorption on surfaces from either the gas phase or the liquid phase under conditions of controlled temperature, relative humidity and flow velocity. Measurements have been made of iodine absorption rates on paints and metal surfaces. Samples of materials used in testing for the sump blockage issue have been provided to the investigators. Tests have shown there to be little tendency for iodine gas to absorb on reflective metal insulation (RMI). There is a tendency for iodide and iodate ions dissolved in water to absorb on fibrous insulation at acid pH levels (pH= 5). Specimens loaded with iodine in the BIP project can be irradiated for prolonged periods in a closed cell with aliquot sampling. Testing in BIP has been done with the Ripolin epoxy paint and with Amerlock epoxy paint often found in containments of US reactors.

**Findings**

The Phébus-FP tests have yielded information that confirms the MELCOR modeling used for reactor accident analysis and development of the alternative source term (NUREG-1465). The tests have also yielded new insights listed below.  These insights are being used to modify the MELCOR code and the NUREG-1465 source term as appropriate.

1. Cesium (Cs) is actually released from the overheated reactor fuel as cesium molybdate ($Cs_2MoO_4$) vapor rather than as cesium hydroxide (CsOH) as has been assumed in many

past accident analyses. $Cs_2MoO_4$ is less volatile than CsOH and is more prone to condense on surfaces in the reactor coolant system (RCS). CsOH that does reach the containment originates from the revaporization in high temperature steam of cesium molybdate deposited in the model of the RCS. Materials including cesium that are transported from the RCS into the containment sump affect the pH of the sump waters. The pH of the sump waters, in turn, affects the partitioning of dissolved iodine from the water into the containment atmosphere as a gaseous species. Hydrolysis of $Cs_2MoO_4$ in the sump is much less effective at keeping the sump waters at high pH than is CsOH. Consequently, severe accident analyses need to reflect this change in the dominant form of cesium that passes through the RCS to the containment. Because of the significant release of molybdenum relative to other members of the refractory metal class of radionuclides (Ru, Pd, Rh, etc.), it is advisable to create an additional radionuclide class for molybdenum.

2. Iodine released from the overheated reactor fuel transport in both gaseous and particulate forms. CsI is among the chemical forms of particulate iodine. However, tests (FPT-0, FPT-1, and FPT-2) with silver-indium-cadmium control rods suggest that cadmium iodide ($CdI_2$) is a more common form of the particulate iodine.

3. Particulate materials are transported through the RCS and containment as agglomerates of mixed substances rather than pure substances. Metals from control rod and structural materials, oxides including materials from fuel and cladding, and fission products are found in a typical particle that develops following release of materials from overheated reactor fuel.

4. Silver vaporized from control rod alloys in tests FPT-0, FPT-1 and FPT-2 passed through the RCS into the containment sump where it reacted with any dissolved iodine species to form insoluble AgI and $AgIO_3$. These reactions of silver limit the concentration of dissolved iodine in sump waters to very low levels. Such low concentrations did not support substantial partitioning of iodine from the sump into the Phébus containment atmosphere regardless of the sump pH.

5. Tellurium is not extensively sequestered in unreacted zirconium alloy cladding on the fuel during fuel degradation. Tellurium is instead extensively released from the overheated fuel. It adopts a chemical form that does not react extensively with the nickel alloy tubing that makes up the RCS model in the Phébus tests. Any deposition in the RCS model is due largely to the deposition of aerosol particulate. Significant amounts of tellurium were observed to reach the containment model.

6. With respect to the behavior of iodine in containment, the most important observation is that in tests FPT-0, FPT-1, FPT-2, and FPT-3 steady-state iodine concentrations developed in the containment model atmosphere. This steady state concentration persisted for days (up to 96 hours). (Test FPT-4 was a test with a debris bed of irradiated fuel and oxidized cladding that could not be re-irradiated to build an inventory of short half-life iodine.) The steady state concentration of gaseous iodine in the containment atmosphere developed whether the containment sump water was basic (pH=9) or acid (pH=5). It is likely that iodine that did reach the containment sump had no involvement in the development of the gaseous iodine concentration in the sump. In tests with silver-indium-cadmium control rods (FPT-0, FPT-1, FPT-2), iodine in the sump either precipitated as a silver salt (FPT-0 and FPT-1) or was effectively sequestered as a result of high pH (FPT-2). Indeed, it was observed that the steady state concentration of iodine in the containment atmosphere decreased when the sump water temperature was increased so that the sump waters evaporated and increased when the sump water temperature was adjusted to promote condensation of steam in the

sump – precisely the opposite behavior of what would be expected if iodine were partitioning from the sump into the containment atmosphere.

The constancy of the gaseous iodine concentration in the containment atmosphere suggests that there are continuing sources and sinks for iodine in the containment. The steady state concentration is the concentration at which the source rates and the removal rates for gaseous iodine balance. Observations made during the tests as discussed above show that the sources and sinks are independent of the containment sump. Empirical observations during the tests and observations of surface contaminations suggest that iodine was interacting with the painted surfaces of condensers in the Phébus containment. These condensers were incorporated into the design to simulate the surfaces of structures and equipment that will be present in reactor containments. The pH of water films that form on the surfaces by condensation are unaffected by buffers placed in the sump. The water films may be acidified in fact by the radiolysis of air to form nitric acid or the radiolysis of cable insulation to form hydrochloric acid and sulfuric acid. In the acidic medium, dissolved iodine particulate will be oxidized by radiolysis to form molecular iodine. It has long been known that dissolved molecular iodine or gaseous molecular iodine will absorb into paint. The absorption is not readily reversed simply by heating the paint.

7. Both the BIP and EPICUR tests have confirmed that gaseous molecular iodine absorbs on paint. They have also shown that molecular iodine dissolved in water will absorb into immersed paint surfaces. EPICUR tests have shown that irradiation of iodide solutions produces a chemical form of iodine – probably molecular iodine – that readily absorbs into paint. Both EPICUR and BIP have shown by different means that iodide in solutions that are not irradiated will little absorb into paint.

   EPICUR tests have shown that irradiation of paint contaminated with absorbed iodine leads to the evolution of small amounts of particulate iodine and more important amounts of molecular iodine and volatile organic iodide. Tests done in support of the EPICUR program have shown that molecular iodine and volatile organic iodides undergo decomposition due to radiolytic and thermal processes to form very small particles of iodine oxide (IOx). The IOx particles nucleate with diameters near 0.01 µm and grow to about 0.1 µm in the absence of extensive amounts of additional aerosol. These are particle sizes known to be difficult to filter or to be removed by suppression pools. Such small particles do not sediment at rates meaningful for reactor safety considerations. The particles can diffuse and be convected to surfaces. If water films are present they will dissolve and if irradiated reabsorb into paint. The cycle of absorption, irradiated desorption, destruction in the gas phase and redeposition can repeat to produce a steady state concentration of iodine and a concentration of very fine iodine particulate in the containment atmosphere.

8. BIP experiments of iodine absorption onto paint have shown that the ambient partial pressure of steam is as important as temperature in the rate of iodine absorption. BIP tests have also shown that there is little tendency for gaseous iodine to absorb on prototypic samples of reflective metal insulation (RMI). There is, on the other hand, a tendency for iodate ions in solution to absorb onto fibrous insulation under acid (pH=5) conditions. Tests are underway to see if dissolved iodine ions will absorb on goethite and aluminum hydroxide expected to be present in containment sumps. No important tendency for iodide ions dissolved in water to absorb on paint was detected. Oxidation of iodide to molecular iodine in solution led to rapid absorption into immersed paint.

9. Testing has been done on two important classes of paints found in reactor containments. Ripolin is an amide cured epoxy paint. Amerlock is an amine cured epoxy paint. There has been more limited testing of a British epoxy paint and a German epoxy paint as well as some testing of a polyurethane paint. The current separate effects test programs are adequately exploring the effects of temperature, steam partial pressure and surface loading on the rates of iodine absorption and radiolytic desorption. Insufficient attention has been given to the effects of paint aging both during normal plant operations and aging produced by accident events. NRC is encouraging the separate effects programs to more systematically investigate aging using guidance criteria used for qualified coatings in reactor containments. There has also been insufficient attention given to the effects of dose rate on observations. Planned upgrades to test facilities will lead eventually to information on the effects of dose rate.

**Application to Severe Accident Modeling**

The Phébus-FP results have been used to upgrade the modeling in the MELCOR accident analysis computer code. Test data have been used to modify modeling of degraded fuel relocation within the core region. For some important accident sequences, these modifications together with improved convective and radiative heat transfer modeling have prolonged the predicted duration of in-vessel core degradation for some accident sequences. The longer duration of core degradation can lead to more extensive release of volatile radionuclides from the reactor fuel. The modeling of cesium release from degrading fuel as cesium molybdate rather than cesium hydroxide has been implemented. For accident sequences that do not involve high flow rates through the RCS, the updated code predicts more extensive cesium deposition in the reactor coolant system and, consequently, less cesium release to the containment. The modeling of tellurium release from degraded fuel and transport of tellurium in the reactor coolant system has been upgraded to better reflect the observations in the Phébus-FP experiments. These modifications were initially tested in MELCOR 1.8.5 and then implemented into MELCOR 1.8.6 and subsequent MELCOR versions. A revision to the alternative accident source term (NUREG-1465) is being prepared based on these improvements to the MELCOR computer model.

Although these changes impact the predicted source term in accidents, the impact may not necessarily be consistent across scenarios. Changes to a less volatile chemical form such as the change of the expected chemical form of Cs from CsOH to $Cs_2MoO_4$ tend to reduce the source term due to enhanced deposition on surfaces leaving less fission product material to reach the containment or the environment. Changes that result in a larger release of a fission product such as the finding that Te was not trapped in unreacted cladding tend to increase the source term as more of this fission product is made available for release to the containment and the environment. At this stage the impact of iodine modifications on the source term cannot be determined because the Phébus-FP experiments were not scaled for containment chemistry. The multiple reactions that produce gaseous iodine or convert it to different forms depend on several parameters that differ between actual reactor containments and the Phébus-FP experiments. The differences include surface-area-to-volume ratios, type of paint, and the potential for competitive reactions with compounds released from cable degradation in actual reactor containments that were not present in the experiments. These effects need to be properly scaled. The modeling currently being developed to perform this scaling will be implemented in MELCOR following testing.

The observations concerning gaseous iodine made in the Phébus-FP experiments depart significantly from what was expected based on the paradigm adopted in the past. The tests have confirmed that iodine is released to the containment primarily as aerosol particulate with a small fraction of iodine gaseous form. It had been assumed that particulate iodine would deposit within containment by gravitational or other means and eventually accumulate as a solute in the containment sumps. Gas iodine would either chemically deposit on surfaces or undergo radiolytic or thermal reactions in the containment atmosphere. The water soluble deposits and decomposition products would also accumulate in the containment sumps. It was assumed that the inventory of gaseous iodine could only be replenished by partitioning of molecular iodine from the sumps to the atmosphere and this could be suppressed by assuring sump waters were basic (pH> 7). When sump waters were kept basic, the concentration of gaseous iodine would fall to levels that were negligible for purposes of safety analysis.

Contrary to the above assumptions, the test data show that very low concentrations of gaseous iodine are not achieved regardless of the sump pH. There is instead an interaction of iodine with painted surfaces within the containment. These interactions lead to a steady-state concentration of gaseous iodine. The safety significance of this steady-state concentration depends on how much iodine is involved in the surface interactions and the magnitude of the steady-state concentration that will develop in reactor containments to balance processes that generate gaseous iodine and processes that decompose gaseous iodine. Data from the tests cannot be used to directly infer these quantities for reactor accidents. In the tests more than 15% of the iodine released to the containment was involved in interactions with the painted surfaces. But, iodine released into the model containment had much greater access to the surfaces of containment sumps in the experiments than it is expected to have in accidents at most reactors. Similarly, dose rates in actual accidents and paint aging in reactor containments need to be considered in scaling the results of the tests to accident conditions.

Modeling currently in the MELCOR computer code does not reflect the complexity of containment chemistry thought to explain observations of iodine behavior made during the Phébus-FP tests. The current modeling was based on the RTF tests conducted many years ago in Canada. Efforts are underway to upgrade this modeling. A ‘stand-alone,’ detailed model of iodine chemistry is being developed. This development is supported by the separate effects test programs EPICUR and BIP. The stand-alone model will be used to identify crucial phenomena that need to be incorporated into the systems-level code, MELCOR.

**<u>Application to Accident Source Term</u>**

Results of the Phébus-FP tests have confirmed many of the important features of the NRC’s revised accident source term (NUREG-1465). They have suggested, however, some modifications including separation of the refractory metal radionuclide group into two groups – one composed of molybdenum which is released with cesium following temperature excursions of the core driven by steam interactions with the zirconium alloy cladding on fuel, and a second composed of ruthenium, palladium, rhodium etc. that are not released extensively. Releases of tellurium are also thought to be more extensive during high temperature core degradation because the tellurium is not efficiently sequestered in unreacted zirconium cladding. The integral tests with conventional silver-indium-cadmium control rods did show that most of the iodine released from degrading fuel to the containment model was in the form of metal-iodide aerosol particulate. Typically, less than five percent of the released iodine was in gaseous form.

The final test included a boron carbide control rod rather than a silver-indium-cadmium control rod. This control rod is quite unlike a control blade in a boiling water reactor. A control blade has sufficient stainless steel cladding to dissolve all the boron carbide within the blade so the boron carbide is not exposed directly to steam during core degradation. The thinner cladding on the control rod used for the test incompletely dissolves the boron carbide so the control material will react with steam to form volatile boric acid during core degradation. The boric acid is volatile and is thought to have reacted in the FPT-3 test with other vaporized materials to form stable borates and to cause release of much (~80%) of the iodine to the containment in gaseous form. The test is not thought to be representative of iodine release during core degradation of a boiling water reactor, but does indicate that competitive chemistry in the reactor coolant system can affect the fraction of iodine released to the containment in gaseous form. This result may have implications in the safety analysis of reactor designs that use water as a coolant but use novel materials and configurations for reactor control.

## Regulatory Applications

Overall, the Phébus-FP integral test results and associated analyses have contributed to the staff's confidence in the use of the MELCOR code for safety analysis and risk-informed decision-making. The test data have also increased confidence in the utility of the NUREG-1465 source term for use in design-basis dose assessment reviews (i.e., Regulatory Guide 1.183, "Alternative Radiological Source Terms for Evaluating Design-Basis Accidents at Nuclear Power Reactor"). However, the Phébus-FP results produced unexpected findings concerning the utility of sump pH control of gaseous iodine in the containment following a reactor accident.

The Phébus-FP tests indicated no effects of pH control on gaseous iodine in the containment atmosphere. Indeed, silver ions in the sump from releases of control rod materials assured that even under acidic conditions (pH < 7), the dissolved iodine concentrations were too low to sustain any important partitioning of iodine from the sump to the containment atmosphere. These findings may have implications for the sump screen blockage issue (GSI-191). Moreover, on the basis of experiments reported in NUREG/CR-6913,"Chemical Effects Head-Loss Research in Support of Generic Safety Issue 191," interactions of chemicals used to control sump pH and some insulation materials dispersed to the sump following an accident can exacerbate sump screen blockage. While eliminating sump buffer chemicals may be beneficial for sump strainer blockage concerns, chemical-effects testing has not been performed to address un-buffered sump pool environments. Separate effects experiments are underway to identify other interactions of iodine with materials dissolved or suspended in sump waters. Modeling is being developed to better predict how such interactions will affect iodine partitioning from the sump under conditions when there is not pH control of the sump waters.