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THE RELEASE AND TRANSPORT OF FISSION PRODUCT CESIUM
IN THE TMI-2 ACCIDENT*

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ABSTRACT

Approximately 50% of the fission product cesium was released from the overheated UO_2 fuel in the TMI-2 accident. Steam that boiled away from a water pool in the bottom of the reactor vessel transported the released fission products throughout the reactor coolant system (RCS). Some fission products passed directly through a leaking valve with steam and water into the containment structure, but most deposited on dry surfaces inside of the RCS before being dissolved or resuspended when the RCS was refilled with water. A cesium transport model was developed that extended measured cesium in the RCS back to the first day of the accident. The model revealed that ~62% of the released ^{137}Cs deposited on dry surfaces inside of the RCS before being slowly leached and transported out of the RCS in leaked or letdown water. The leach rates from the model agreed reasonably well with those measured in the laboratory. The chemical behavior of cesium in the TMI-2 accident agreed with that observed in fission product release tests at Oak Ridge National Laboratory (ORNL).

INTRODUCTION

The TMI-2 nuclear reactor accident occurred on March 28, 1979. Many months passed before an accurate measurement of fission product release could be made because high radiation levels prevented access to many locations where fission products were deposited. The mass balance for ^{137}Cs for December 31, 1979 (278 d after the accident), showed that 44.6% was located outside of the RCS, the high-pressure, primary circulating system.¹ This is equivalent to 375 KC1 of ^{137}Cs decay corrected to the day of the accident.

In order to help determine the nature and amount of fission product migration throughout the reactor system during the first days of the accident, a cesium transport model was developed. The model, described in detail later in the text, used a trial-and-error procedure which combined assumed rates of cesium transport to the RCS water with rates of known water leakage. A good fit was obtained in which the transport model matched even the earliest (second day) measurements of cesium concentration in the RCS water.

There is no way to determine the chemical forms of cesium that existed at the time of release and transport during the TMI-2 accident; however, there is good reason to believe that the chemical behavior of cesium at TMI-2 was the same as that observed in the ORNL fission product release tests. In the ORNL tests, the behavior of cesium in a steam-helium-hydrogen atmosphere was that of CsOH and CsI .² Essentially all of the released iodine behaved like CsI , accounting for ~10% of the released cesium and 99.5% of the released iodine; the remaining 90% of the cesium behaved like CsOH . The CsI proved to be very stable in these tests even though it was exposed to many potentially reactive

surfaces in the apparatus. Cesium hydroxide, on the other hand, was very reactive; it reacted with and was retained by several surfaces, such as SiO_2 , ZrO_2 , and oxidized stainless steel. The resulting cesium compounds were less volatile than CsOH , and much of the cesium was retained at higher temperatures. Like CsI and CsOH , these compounds were found to be soluble in simulated TMI-2 RCS water but dissolved more slowly. The cesium transport model revealed an essentially identical leaching pattern for the ^{137}Cs in the TMI-2 RCS, as was obtained by leaching cesium from stainless steel and zirconia surfaces. Details of cesium chemical behavior as observed at ORNL follow in addition to a description of a leachability study.

CHEMICAL BEHAVIOR OF RELEASED CESIUM IN THE ORNL FISSION PRODUCT RELEASE TESTS

Background of Cesium Chemical Behavior

Since 1975 a total of 28 fission product release tests have been conducted at ORNL for the U.S. Nuclear Regulatory Commission (NRC).³⁻¹¹ Highly irradiated fuel segments, cut from fuel rods from six different commercial light-water reactors (LWRs), were heated in the temperature range 500 to 2000°C. Three different test atmospheres were examined: two tests in dry air, two tests in inert gas, and twenty-four tests in steam-helium-hydrogen. The release behavior of cesium under simulated, severe accident conditions has been well characterized and documented.

The experimental apparatus consisted of a furnace assembly, in which the fuel rod segments were heated, and a fission product collection system. During a test, the volatile fission products were released from the fuel segment into the flowing carrier gas and transported from the furnace tube to the collection system. The first stage of the collection system was a platinum-lined thermal gradient tube (TGT) where the volatile species deposited according to their dew points. Downstream from the TGT was a filter pack that collected particulates and the more volatile and penetrating species, such as I_2 , HI , and CH_3I . The analytical methods employed in these studies were gamma spectrometry, neutron activation analysis, spark-source mass spectrometry, X-ray diffraction, and energy dispersive X-ray analysis.

Experimental Results

The observed release behavior of cesium in the 24 steam-helium-hydrogen atmosphere tests was that of CsOH and CsI . The release fractions of cesium and iodine inventories varied from a low of 0.00004% (0.02 μg) and 0.0002% (0.1 μg), respectively, to a high of 59% (133 mg) and 53% (13.6 mg), respectively. The released iodine form, believed to be CsI , proved to be very stable in the gas phase: it showed little tendency to react with or be decomposed by hot quartz, hot zirconia, and hot oxidized and unoxidized stainless steel surfaces that it contacted. Several tests clearly demonstrated that the iodine was already in the CsI form upon exiting the fuel segments, at least at temperatures up to 1600°C. The cesium that was not associated with iodine and presumed to be CsOH was chemically very reactive. Its collection behavior showed that (1) it was almost quantitatively gettered and retained by quartz (SiO_2) surfaces of the apparatus at temperatures $<950^\circ\text{C}$ (probably forming cesium silicate), (2) it was significantly gettered and retained by zirconia in the temperature range 800 to 1200°C (probably forming cesium zirconate),

and (3) it was retained by oxidized stainless steel at measured temperatures between 600 and 850°C (probably forming cesium chromate and silicate).²

The TGT was used as a collection device to characterize fission products according to their volatility by observing the temperature at which they deposited on the wall of the tube.¹²⁻¹³ The deposition peak for a fission product species occurs at its dew point, which depends upon the vapor concentration of the species. To aid in the interpretation, deposition patterns of known compounds were measured in control tests and were compared with those of unknown fission product compounds in the fuel tests.

The TGT deposition profiles for iodine in the six HI tests are shown in Fig. 1. Since there was only one iodine peak in each profile, it is highly likely that there was only one form of iodine that deposited. The temperature range in which the maximum surface concentration occurred for each iodine peak was between 575 and 410°C. In tests HI-3 through -6, in which maximum surface concentrations were similar (310, 265, 300, and 350 $\mu\text{g}/\text{cm}^2$, respectively), the temperature range of the peak concentrations was relatively narrow, 530 to 500°C. The deposition behavior for iodine as CsI in the control tests, as well as in the implant tests, was similar to that obtained for iodine in all the irradiated fuel tests. The peak temperature range in these tests was 570 to 360°C.

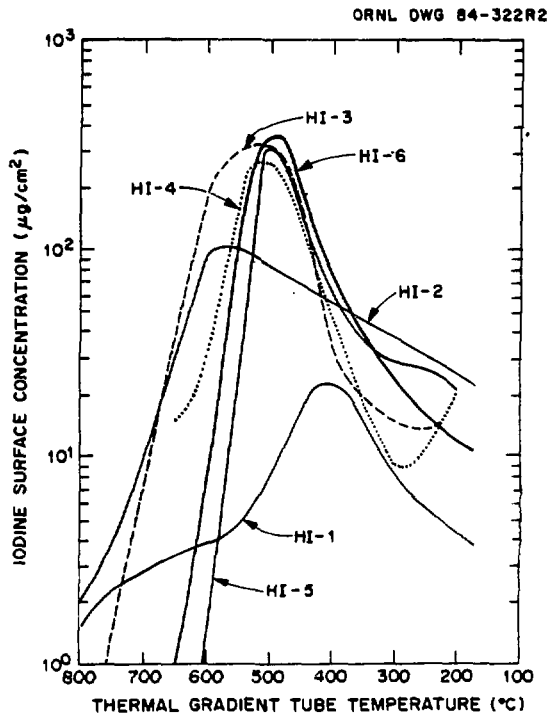


Fig. 1. Deposition of iodine in tests HI-1 through HI-6.

In test HT-2,⁴ in which an H. B. Robinson-2 fuel segment was heated to 1450°C, almost all the released cesium except that associated with iodine was rapidly gettered and retained by the quartz surfaces in the furnace tube near

the defect opening in the cladding of the fuel segment. The TGT vapor deposition profiles for iodine and cesium are illustrated in Fig. 2. Considering the facts that (1) almost equal quantities of cesium and iodine were found on the liner of the TGT, (2) both elements deposited in the same peak region at $\sim 500^\circ\text{C}$, and (3) the peak location was similar to those obtained for CsI in the control tests, it is apparent that the iodine and cesium were deposited in the TGT as CsI. (Most of the released iodine, 90%, was found in the TGT; only 0.06% was found on the charcoal as elemental iodine.) Furthermore, the collection behavior in this test clearly showed that the iodine was already in the CsI form or CsI was formed instantly upon exiting the segment. Had the released cesium and iodine been in the elemental form on exiting the segments, all the cesium would have been gettered by the quartz and all the iodine would have transported to the charcoal.

In one of the recently conducted HI tests (HI-6),¹¹ a stainless steel rather than a platinum TGT liner was used. Gamma spectrometric analysis showed that the major portion of the cesium was deposited between ~ 880 and 600°C at the inlet end of the TGT, as illustrated in Fig. 3, the region where surface oxidation occurred. The degree of oxidation was determined to be more severe at the higher temperature, with the average thickness of the oxide calculated to be $\sim 12\ \mu\text{m}$. It is assumed that the retention of cesium by the oxide layer was the result of its reaction as CsOH with the oxides of chromium, iron, and silicon. Certain experiments have indicated that cesium preferentially reacted with SiO_2 in the oxide layer, forming the silicate $\text{Cs}_2\text{Si}_4\text{O}_9$.¹⁴ Other experiments indicated that the cesium reacted to form cesium chromate.¹⁵

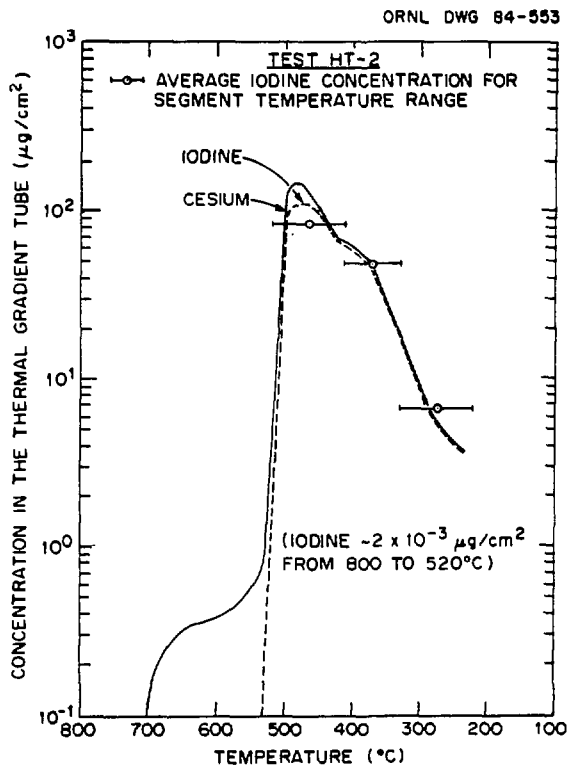


Fig. 2. Cesium and iodine deposition profiles in the TGT of test HI-2.

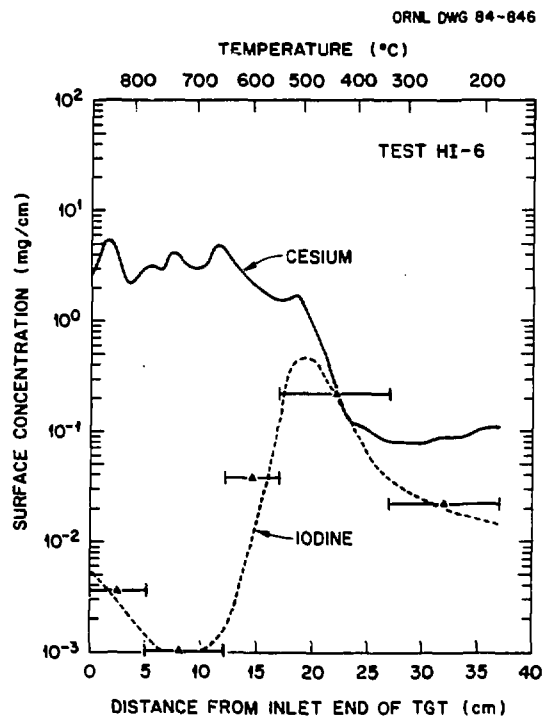


Fig. 3. Cesium and iodine deposition in the TGT of test HI-6.

Calculations indicate that cesium ferrite is also possible.¹⁵ Considering that the average concentration of cesium in the oxide layer of the TGT in HI-6 was ~ 2 mg/cm², and assuming that the silicon in the stainless steel was $\sim 0.5\%$, calculations showed that the cesium/silicon mol ratio in the oxide layer was ~ 12 . This is significantly more cesium (24 times) than is needed to react with the available silicon as SiO₂ in the oxide to form (Cs₂Si₄O₉). These results demonstrated that released cesium reacted with the other oxides as well. More importantly, from the standpoint of reactor safety, the released cesium was significantly retained by oxidized stainless steel at relatively high temperatures. The iodine profile in Fig. 3 was typically one of CsI deposition. The cesium released as CsI proved to be very stable in the gas phase while passing through the oxidized region of the stainless steel TGT.

Leachability of Cesium from Stainless Steel and Zirconia Surfaces

A study was conducted to examine the leachability of cesium from two sections of the stainless steel TGT used in test HI-6, which was discussed above. The sections leached were taken from the oxidized region at the upstream end of the TGT liner (Fig. 3). It should be noted that test HI-6 did not include vapors from either control rod material (i.e., Ag, In, Cd) or from borated water. Section 1, which was 6 cm long, was taken from the hottest region between 880 and 800°C; section 3 came from a cooler region located 12 to 16.9 cm from the entrance, where the temperature was between 675 to 550°C. The leachability of cesium from a section of zirconia ceramic used in control test C-8 was also examined.² In test C-8, vaporized CsOH (traced with ¹³⁷Cs) was quantitatively getterred by the walls of the zirconia furnace tube liner at temperatures between 800 and 1000°C.

To make the study meaningful, a stock leach solution was prepared that simulated TMI-2 cooling water or water that might be found following any pressurized-water reactor (PWR) accident. It was made by adding boric acid and sodium hydroxide in ultra-pure water to provide a solution which contained ~ 3750 ppm boron and 1000 ppm sodium at a pH of 7.8.

Each item was leached in 200 mL (initially) of the leachant for 23.5 h at 53°C. (The cooling water at TMI-2 was initially at a much higher temperature than this, and its temperature decreased following the accident.) One-milliliter samples were analyzed periodically to follow the leaching rate. Each leach was stirred in the same manner.

Figure 4 shows the percentages of deposited ¹³⁴Cs removed from the oxidized stainless steel and ZrO₂ sections with time. Cesium was more readily removed from section 3 than section 1 of the stainless steel TGT used in test HI-6. After a 15-min leaching period, 67% of the cesium had been removed from section 3 compared with 35% from section 1. After 23.5 h, 99.5% had been removed from section 3 and 90% from section 1. These data show that the reaction products of cesium and the oxidized stainless steel components, when formed in steam at higher temperatures, are more difficult to remove with PWR cooling water at 53°C than when formed in steam at lower temperatures. At the higher temperatures, a larger fraction of the cesium is apparently more deeply embedded in the oxide layer, which was also thicker. A portion of the cesium at the lower temperature may have condensed rather than reacted with the stainless steel surface. If so, this may account for the faster leach rate for section 3. Species of cesium like CsI and CsOH are readily soluble in water.

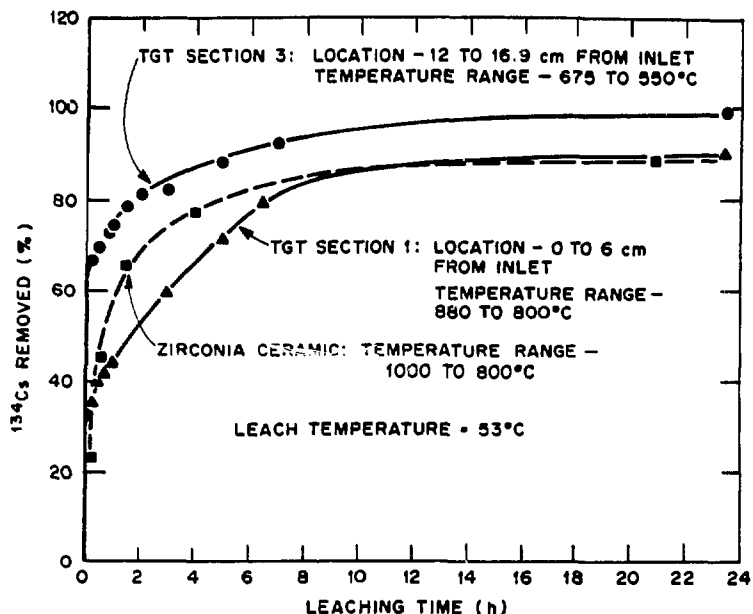


Fig. 4. Leaching of cesium in TMI water.

The leach behavior of the cesium which reacted with and was retained by the zirconia ceramic was similar to that of the cesium on section 1 of the stainless steel TGT liner (see Fig. 4); only 90% was removed after 1 d of leaching. After similar leaching was continued for 8 d, an additional 6% was removed. Apparently, there was a portion of the reacted cesium that was deeply embedded that resisted leaching.

DEVELOPMENT OF THE CESIUM TRANSPORT MODEL

Figure 5 shows the condition of the TMI-2 reactor at the time of the first major fission product release from the fuel.^{17,18} The water remaining in the RCS was in four pockets: the bottom of the reactor vessel, the bottoms of the two steam generators, and in the pressurizer. Heat from the radioactive fission products caused the water in the reactor vessel to boil, thus transporting fission products throughout the RCS with the flowing steam. Hydrogen was also present as the steam reacted with the Zircaloy cladding.

The fission products were transported as vapor and aerosols and condensed or deposited on the cooler metal surfaces, such as the plenum (the upper part of the reactor), the piping, and the heat exchangers. Some fission products entered the pools of water directly with the condensing steam. The valves at the top of the pressurizer were open during part of the time that the fission products were being released from the overheated core. This allowed steam and hydrogen to carry release fission products into the pressurizer where flashing liquid carried them out of the RCS. The escaping steam, water, hydrogen, and fission products flowed down through a tank and into the containment building (the main building structure that surrounds the reactor system). Most of this material flowed rapidly to the basement area, but some was carried to the interior surfaces of the containment building. At later times, water was removed from the RCS by means of the letdown system.

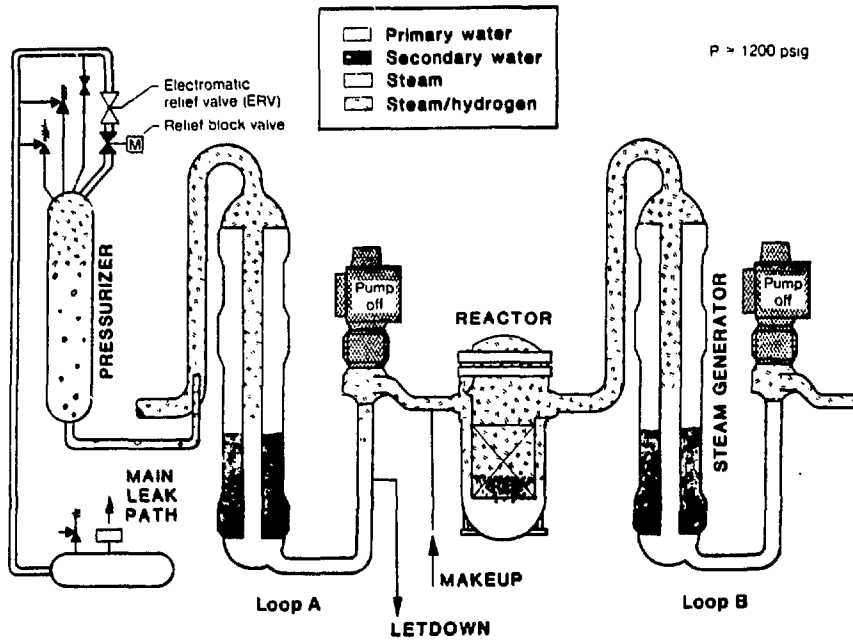


Fig. 5. TMI-2 at time of maximum fission product release.

We assume in the model that all of the cesium leaving the RCS was dissolved or present as fine particles in water that leaked from the RCS. There was no separate leakage of gas or dry aerosol. The refilling of the RCS with water is shown in Fig. 6. The time reference is the start of significant fission product release, 0648 on March 28, 1979. We assumed that the circulating portion of the RCS, 77,000 gal (291.5 m³), was well mixed and that water and fission products entering the pressurizer were immediately leaked to the containment building.

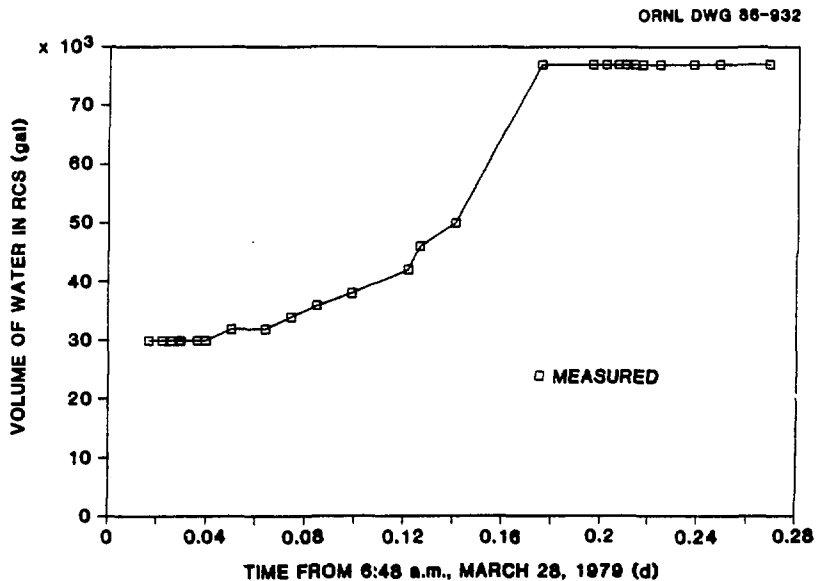


Fig. 6. Volume of water in the reactor coolant system.

We obtained the water leakage and letdown rates from the work of others.^{1,19,20} From these data, as shown in Figs. 7 and 8, it can be seen that there is a difference in the leakage rates of a factor of 1000. Makeup water was assumed to be clean (none of the fission product cesium was recycled through the letdown-makeup system).

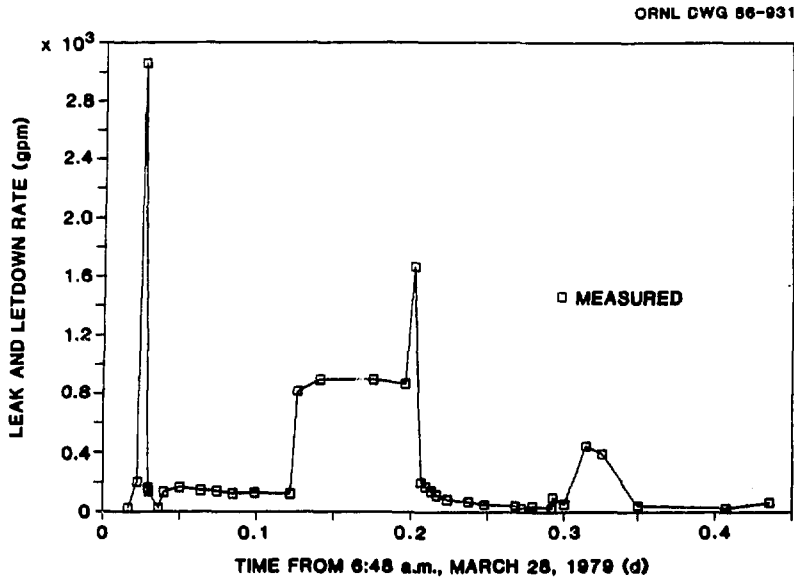


Fig. 7. Water leak and letdown from the RCS.

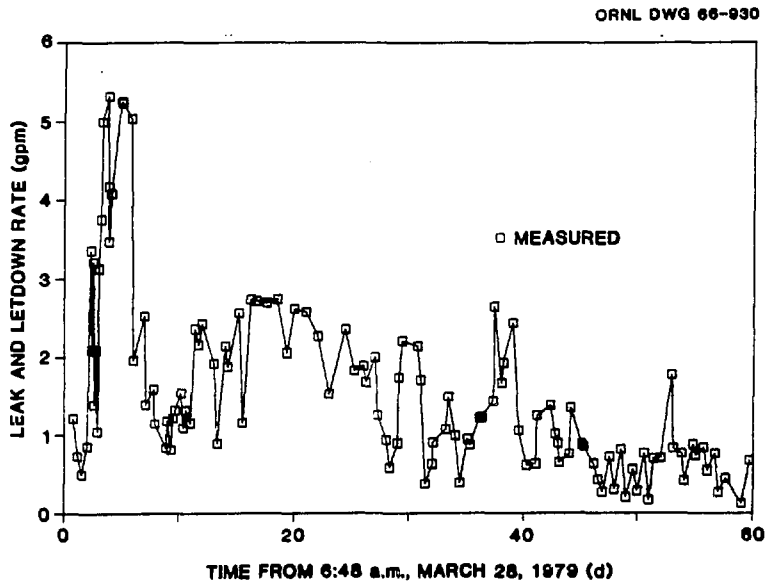


Fig. 8. Water leak and letdown from the RCS.

The only unknown was the rate of transport of cesium into the RCS water. Initially, this was mainly by direct transport from the overheated fuel to the RCS water. Fuel temperatures during the accident were not well enough known to use any of the fission product release models, so we decided to try using assumed release rates and times and compare the resulting RCS water concentrations with those measured following the accident. The first measurement was performed the day following the accident.

Trials with assumed release rates showed that there was a long slow release that continued for days and even months after the accident. As described above, we assumed that cesium was being leached from the fuel and other material in the RCS. Using a trial-and-error process, we adjusted the direct release rates and the slower leach rates until a good match was obtained for measured quantities of cesium in the RCS. The amount of cesium found in the pool of water in the basement was also used as a target for the model even though it was not measured until 155 d after the accident.

RESULTS OF THE CESIUM TRANSPORT MODEL

A good fit with the cesium contents measured in the RCS water and the basement pool was a release of 50 KCi/h for 2.4 h (120 KCi total) transported directly to the RCS water followed by the leach rate labeled "Model," which is shown in Fig. 9. The horizontal dotted line represents cesium transported directly from the fuel to the RCS water, while the solid sloping line depicts the leaching of cesium. The changeover undoubtedly did not occur as abruptly as shown.

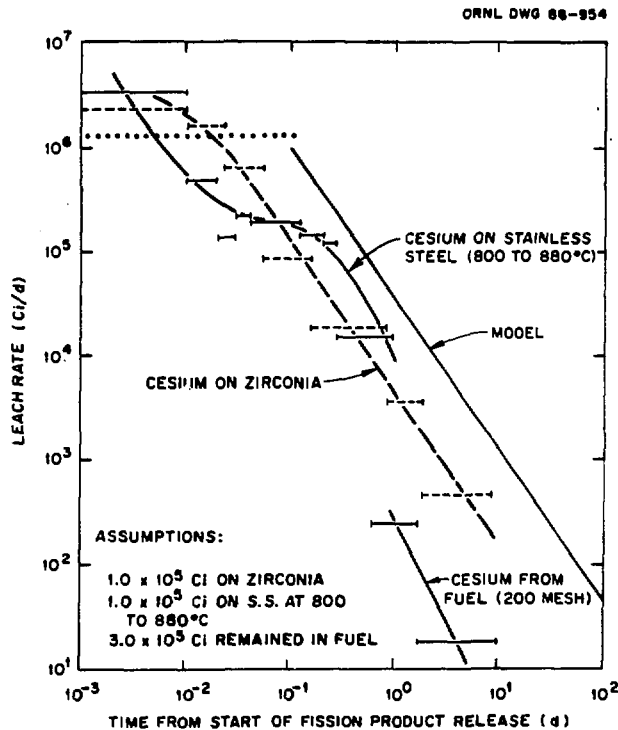


Fig. 9. Leaching rates scaled to TMI-2.

Three other lines are also shown in Fig. 9, illustrating laboratory results for the leaching of cesium from ZrO_2 , oxidized stainless steel (see Fig. 4), and crushed, irradiated (but never overheated) UO_2 fuel.²¹ The leach rates are scaled up to what might have existed in TMI-2. There is a definite similarity between the laboratory results for leaching and the best fit for leaching in the cesium transport model. The leaching of cesium from the overheated fuel in TMI-2 was probably greater than from the crushed fuel in the laboratory study. According to the model, the leached component amounted to ~231 KCi by December 31, 1979. Only 14.8 KCi remained in the RCS water at that time. The amount of cesium deposited on fixed surfaces at the time of the accident was the same 231 KCi, or 62% of the cesium released from the fuel.

The amount of cesium in the RCS water for 40 d following the accident is shown in Fig. 10, both for the transport model results and for measured values. The agreement is good even out to 278 d (December 31, 1979). Details of the comparison for the first 3 d are shown in Fig. 11. The initial straight-line increase to 120 KCi is the result of the assumed direct transport of cesium released from the fuel. All remaining increases result from leaching of cesium from various surfaces. The rapid drop to 53 KCi is the result of a large amount of water and steam vented through the pressure relief valves. Most other water letdown or leakage was much slower. After 3 d, the amount and concentration of ^{137}Cs in the RCS water decreased because of continued slow leakage of contaminated water, makeup with clean water, and decreasing leach rates.

The total amount of ^{137}Cs leaked from the RCS is shown in Fig. 12. Half of the cesium transported from the RCS came out in the first half day. The total removed in 278 d (December 31, 1979) was 375 KCi.

SIGNIFICANCE OF THE MODEL RESULTS

A sizable amount of the released cesium, 62%, was deposited on fixed surfaces and then slowly leached. This amount of cesium retained on the primary system surfaces is equal to or greater than that calculated by the NRC's Source Term Code Package (STCP).²² Agreement with this part of the STCP was not an objective of the cesium transport model.

ACCURACY OF THE TRANSPORT MODEL

There is a considerable uncertainty associated with the early hours of the model. Different combinations of direct release and leaching during the first 0.2 d (4.8 h) could produce the same results. The average for this time period must be maintained, however.

One of the biggest uncertainties is the water leakage rate early in the accident. An important contributor to cesium purged from the RCS is the water released by the pressure relief valves (safeties) between 0.12 and 0.2 d. The large amount of cesium released at this time can be seen in Fig. 11. It is not certain that the system pressure rose sufficiently to cause safety relief valve operation. If this water flow is eliminated, it is necessary to assume a very high rate of transport of cesium from fuel to RCS water at an early time, such as 0.01 d.

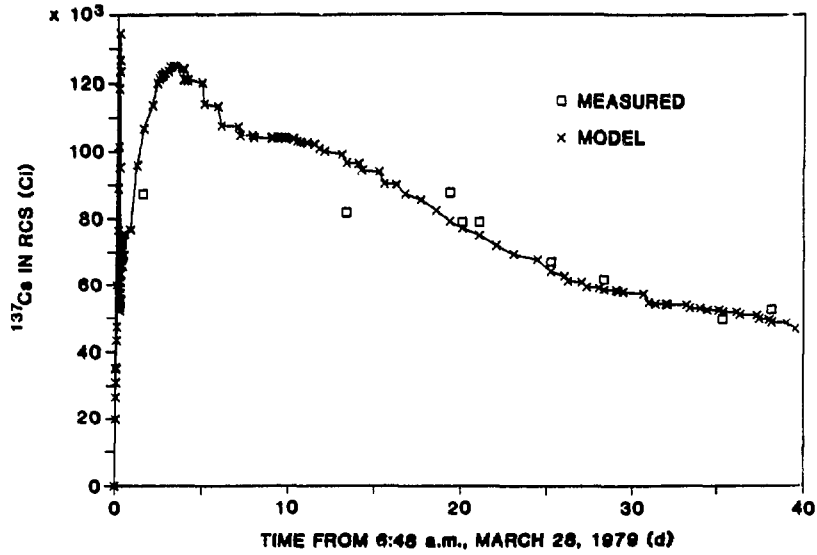


Fig. 10. Amount of ^{137}Cs in the RCS water.

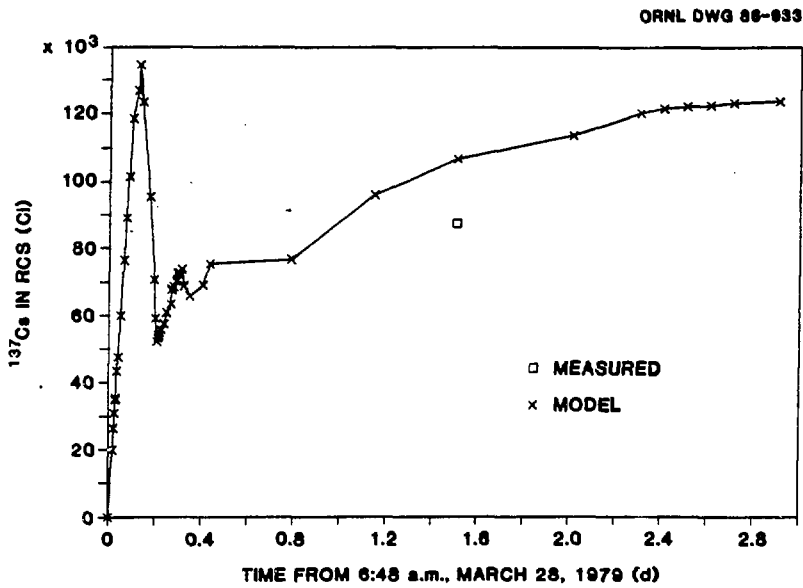


Fig. 11. Amount of ^{137}Cs in the RCS water.

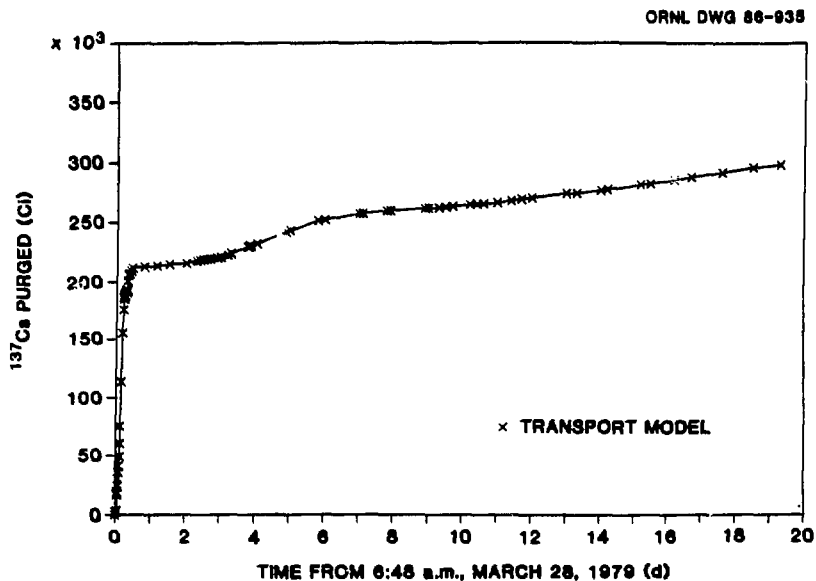


Fig. 12. Amount of ^{137}Cs purged from the RCS.

The leach rate in the model is considered to be most accurate between 0.54 d (13 h) and 100 d. By 13 h, the water leakage and makeup rates were more accurately recorded, an essential component of the transport model. After 100 d, the leach rate was low compared with the RCS water inventory, 50 Ci/d compared with an RCS water inventory of 26,000 Ci.

According to the transport model, half of the leachable cesium was removed by 0.35 d (8.4 h). The earliest amount leached cannot be definitely distinguished from cesium that might have transported directly to the RCS water. The leachable cesium, which deposited on fixed surfaces, could be more or less than the 62% given by the model. The range could well be 50 to 75%. The amount of cesium deposited on surfaces in TMI-2 may be higher than in many other sequences because it was essentially a closed system, while the fuel was at its highest temperatures. A reasonable comparison with experiments is the ORNL test HI-6 in which fission product cesium released from a segment of an overheated irradiated fuel passed through a stainless steel TGT before being collected on filters. Seventy-five percent of the released cesium deposited on (or reacted with) zirconia furnace components at 900 to 1200°C and the oxidized stainless steel TGT at 600 to 900°C; only 25% reached the filters.

The presence of a strong leaching component in the cesium transport model is in opposition to a statement in ref. 1, p. ix. Those authors state that "the data suggest that tritium and cesium were not leached from primary solids and surfaces after the accident." They did not calculate the effect of leakage or makeup and letdown as we did. We believe they observed that the strontium concentration in the RCS water had increased as the result of a leaching process and that since the cesium behavior was different, it was probably not being leached from RCS surfaces.

CONCLUSIONS

1. According to the cesium transport model, ~60% of the fission product cesium released from the fuel deposited on fixed surfaces of the RCS and was slowly leached by the RCS water. This agrees with the NRC's Source Term Code Package (STCP), which shows that in many accident scenarios, a large fraction of the cesium reacts with or otherwise deposits on the primary system surfaces.
2. The leaching behavior in the cesium transport model is consistent with that observed in the laboratory for cesium reacted with zirconia and oxidized stainless steel.
3. The leaching behavior for cesium in TMI-2 is in direct disagreement with one of the conclusions in GEND-INF-047.²⁰
4. The concentration and amount of ^{137}Cs in the RCS water fluctuated considerably during the first 7 h following fuel rod failure because of irregular water leakage (letdown) and rapid leaching rates.
5. The greatest uncertainty in the transport model is in the rate of water leakage and letdown.

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