MANAGEMENT OF SOLID RADIOACTIVE WASTES FROM THE TMI-2 ACCIDENT

A REGULATORY PERSPECTIVE

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ABSTRACT

The TMI-2 accident has resulted in production of unique radioactive waste materials never previously encountered by a commercial nuclear power plant licensee. These materials are of high specific activity, with long-lived isotopes primarily Cs-137 and Sr-90. In addition some TRU contaminated wastes were produced. Federal government assistance was required to facilitate the licensee's processing some of these materials and arranging for satisfactory offsite disposal of all of these abnormal waste forms.

BACKGROUND

Following stabilization of the Three Mile Island Unit-2 (TMI-2) plant after the accident in March 1979, the Nuclear Regulatory Commission (NRC) was faced with unprecedented regulatory, licensing and policy issues for cleanup activities. In its oversight of the cleanup NRC has utilized, to the maximum possible extent, the existing regulatory framework. However, in carrying out its mandate for maintaining safety for the work force and the public, and protecting the environment during the cleanup, NRC was confronted with a number of unique regulatory concerns. Many of these concerns ultimately came down to radioactive waste management issues which were in many ways different from those considered by the NRC for other power reactors. Although the basic technology was available in the National Laboratories, the scale and technical complexity of the required waste management operations (along with decontamination and defueling) had never been undertaken by a U.S. utility company. No nuclear power plant operator had previously faced the problems of processing, handling and packaging of the unique radioactive waste materials onsite, or the safe disposition offsite. As a result, a unique relationship was established among General Public Utilities Nuclear Corp. (the licensee), NRC as the regulator, and the Department of Energy (DOE) involving the expertise existing in the laboratories' contractors. Progress to date in the cleanup (Ref. 1) has benefited from this unique relationship.

NRC's regulatory criteria for reactors are for an operating plant and significant modifications were necessary. This was initially accomplished by modifications made to the facility license in February 1980, with significant changes continuing to be made as the cleanup progresses. TMI-2 might be considered, from a regulatory (Part 50 licensee) perspective, a facility somewhere between a construction project and an operating plant. Another view might be that TMI-2 is more akin to a material license (Part 70) situation. However, no matter which approach is taken (NRC has chosen to retain the Part 50 license - modified for the plant conditions) one must bear in mind that a highly contaminated facility, with a relatively unknown geometry, but significantly damaged core is under the responsibility of an electric utility company.

The most significant policy and licensing issue confronting the NRC concerned the disposition of the unique radioactive solid wastes generated as a result of cleanup activities. NRC established early in the cleanup that the guiding principle would be that the site should not become a long-term radioactive waste repository. The TMI site location on an island in a major river, with a fairly high population density in the area and downstream, clearly precluded this option. It was obvious that decontamination of the facility, including removal of the radioactive wastes and the entire damaged reactor core was necessary to minimize the threat to the health and safety of the public, and to ensure protection of the environment.

Low Specific Activity Wastes

One of the first waste management issues for TMI-2 soon after the accident was the total ban by the Governor of South Carolina from disposal of TMI-2 low level wastes (LLW) at the Barnwell burial site. For the first six months after the March 28, 1979 accident, TMI-2 LLW remained on site until contractual arrangements were made with the U.S. Ecology burial site at Richland, Washington. No satisfactory technical rationale exists for the South Carolina ban, but the result has been added expense to the cleanup and a slight increase in public risk due to the much greater transportation distance. This ban, even for the relatively benign materials involved (trash, anti-C's, etc.) continues to this date, even though TMI-1 materials have been disposed of at Barnwell throughout this period.

High Specific Activity Wastes

At the onset of the cleanup there was no agreed-upon offsite repository for the high specific activity wastes, TRU contaminated materials and the damaged fuel. In spite of this, NRC's position was that removal of these materials was mandatory and we steadfastly maintained that position while seeking a resolution to this TMI-2 problem. This problem was beyond the licensee's capability to resolve. Notwithstanding the unresolved national question for ultimate disposition of high level waste, NRC was eventually successful in formulating an agreement with the DOE for offsite disposition of the entire core and high specific activity wastes for TMI-2 (Ref. 2).

This agreement was not reached however until nearly three years after the accident, and until that time there was always the potential that a successful legal challenge could stop the cleanup. In particular the processing of accident generated water in the containment building could have been delayed. NRC review of the Submerged Demineralizer System (SDS - discussed below) leading to approval was under serious question by a number of groups who raised objections to producing concentrated wastes for which removal from the TMI site was not assured. Only when this agreement for disposition of high specific activity wastes was reached with DOE in July 1981 could the NRC approval action taken in June be implemented without the potential of a serious challenge.

The agreement reached between NRC and DOE covers high specific activity and/or TRU contaminated wastes which are not normally encountered at commercial nuclear power facilities. Waste forms include organic resins from the EPICOR-II System, inorganic zeolites from the SDS, filters and organic resins from the plant Makeup and Purification System demineralizers, other TRU contaminated wastes, and the entire damaged core.

DOE's role as given in this Memorandum of Understanding (MOU) with NRC falls into either of two categories. First, where DOE determines that generically beneficial research, development and testing of the TMI-2 accident generated solid wastes can be carried out, DOE will perform such activities at appropriate DOE facilities. Second, for those other wastes that cannot be disposed of in commercial low level waste facilities because of their abnormal characteristics, DOE may assume responsibility for removal, storage, and disposal to the extent GPUN provides reimbursement to the DOE. In a subsequent paper at this conference, the DOE R&D program for these materials is discussed in more detail (Ref. 3).

A major milestone was reached July 1982 when it was agreed that the entire reactor core would be disposed of by DOE, on a reimbursable basis. Recent actions have been taken since then to clarify provisions of the MOU pertaining to transuranic waste (exclusive of the core). For those accident wastes which after appropriate processing cannot meet 10 CFR 61 criteria for commercial burial due to TRU contamination, DOE will accept these wastes on a reimbursable basis. Acceptance is on the basis that ultimate disposition (after an assumed maximum storage period of 20 years) will be in a Federal repository for those TRU wastes not committed to an R&D program. With these agreements, we have assurance that all of the high specific activity and TRU contaminated wastes will ultimately be disposed of off the TMI-2 reactor site, fully meeting the objectives established by NRC at the start of the TMI-2 cleanup.

Accident Water Processing

Two water processing systems were built after the accident and utilized at TMI-2. Accident water collected in the auxiliary building (<100 μCi/ml) was processed by the EPICOR-II System which utilized organic (or organic plus zeolite) ion exchange media. The first stage liners of this system collected the bulk of the cesium and strontium, and 50 of these liners ended up with specific activities on the average over 30 times higher than normal resin wastes generated at PWR's.

Due to radiolytic degradation of the higher loaded EPICOR-II organic resins, which was not originally anticipated, special tools were developed by DOE to vent hydrogen and inert the resin liners with nitrogen. These 50 liners were then shipped offsite to the DOE laboratory at INEL, Idaho awaiting disposal as Class C waste under 10 CFR 61. Current plans are to dispose of these wastes in high integrity containers at a commercial burial site, except for some wastes to be used in DOE's R&D programs.

Other lower activity resins used in EPICOR-II processing of auxiliary building water have also been shipped offsite. NRC original requirements for solidification of these resins was relaxed to allow disposal in a dewatered form. This action was taken to minimize worker exposures that could result from the complex solidification operation. The balance of the EPICOR-II liners remaining from auxiliary building water processing (3 prefilters - first stage, 15 cation bed liners - second stage, and 7 mixed bed polishing liners - third stage) were disposed of at the bottom of a 9.1 m. (30 ft.) deep slit trench cut into the bottom of an existing commercial burial excavation. The 25 liners were also placed inside covered concrete pipe sections. These extra precautions were taken because of the higher than normal Sr 90 levels in these liners.

The higher level (165 - 180 μCi/ml) water which collected in the reactor building and water in the reactor coolant system was processed through the Submerged Demineralizer System (SDS) (Ref. 4). SDS operation resulted in 19 liners of zeolite, loaded up to 210 Ci/L (6000 Ci/ft^3).

As in the case of the highly loaded EPICOR-II liners, the SDS zeolite liners also produced hydrogen. In the SDS liners, radiolysis of entrained water in the zeolites required the development of a vacuum outgassing system to minimize moisture content. In addition, a gas recombination system utilizing platium/palladium catalysts was used to ensure less than stoichiometric hydrogen-oxygen concentrations during shipping and storage. All 19 of these SDS liners have been shipped to DOE Hanford, Washington facilities for vitrification in DOE's R&D programs or disposal in a monitored burial site.

TRU Wastes

In addition to these high specific activity wastes, estimates have been made of the total volume of TRU contaminated (>100 nci/gm) waste anticipated from the TMI-2 cleanup. It appears that the total volume will be 42 - 57 m^3 (1500 - 2000 ft^3), but considerable uncertainty exists in the concentrations of the remaining reactor building sump sludge. This TRU waste is unique, because of the accompanying Cs 137 and Sr 90 which is not usually encountered when commercial fuels are reprocessed.

Conclusion

The TMI-2 post accident period has been as unprecedented as the accident five years ago. One of the most significant, but unanticipated, results has been the generation of wastes approaching those usually defined as HLW. Without the involvement and cooperation of DOE and NRC, it is clear that a private nuclear plant owner cannot cope with these materials.

TABLE I

Comparison of TMI-2 Accident Water Processing Wastes with LLW and HLW Data (Daughter Products Excluded)

	Reactor Experience	EPICOR-II Wastes	SDS Waste	HLW Characteristics
Bulk Specific Activity, Ci/L	.0035-.35	.14-1.5	53-210	14-35[1]
	(avg. .035)	(avg. >1.3)		8-9[2]
Inventory, Ci	1100[4]	>56,000	440,000	11,000,000[3,4]
TMI-2 Total Volume, liters	---	42,000	3700	---
Significant Radionuclides	Co-58	Cs-134	Cs-134	Cs-137
	Co-60	Cs-137	Cs-137	Sr-90
	Cs-137	Sr-89	Sr-89	Transuranics
		Sr-90	Sr-90	

[1] Idaho HLW Calcines

[2] SRP HLW Sludges

[3] 10 CFR 51.20 - Table S-3

[4] Annually Per Reactor

REFERENCES

1. G. KALMAN and R. WELLER, "Progress in the Recovery Operations at Three Mile Island Unit 2," Nuclear Safety, 25, 1, (1984).

2. Memorandum of Understanding Between the NRC and DOE Concerning the Removal and Disposition of Solid Wastes from Cleanup of the TMI-2, (July 15, 1981, revised March 15, 1982).

3. J. K. REILLY, T. McINTOSH, P. J. GRANT, G. J. QUINN, "Removal, Treatment and R&D of Special Waste at TMI-2," Waste Management 84 Symposium, Tucson (1984).

4. K. J. HOFSTETTER and C. S. HITZ, "Processing of the TMI-2 Reactor Building Sump and the Reactor Coolant System," ANS Winter Meeting, TMI-2 Special Sessions, Washington (1982).

REMOVAL, TREATMENT, AND R&D OF SPECIAL WASTE AT TMI-2

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ABSTRACT

During 1983, the removal and shipment of 50 EPICOR-II waste containers and 14 SDS waste containers were completed marking the removal of approximately 95 percent of the accident-generated waste at the Three Mile Island Unit 2 nuclear power plant. Waste immobilization programs for packaging or solidifying the waste progressed, a system for removing the purification demineralizer resins neared completion, and preparations for receiving special transuranic waste at the Idaho National Engineering Laboratory were initiated.

BACKGROUND

The March 1979 accident at the Three Mile Island Unit 2 (TMI-2) nuclear power plant created, both directly and as the result of subsequent cleanup operations, certain waste forms that are not routinely encountered in normal light water reactor plants. The Department of Energy (DOE) and the Nuclear Regulatory Commission have agreed through a Memorandum of Understanding (MOU) that TMI should not become a long-term repository for radioactive wastes. Under the terms of this MOU, agreement has been reached whereby DOE will accept the fuel from the damaged reactor and certain "special" wastes for R&D and disposition. These special waste materials include:

- Highly loaded (>0.35 Ci/L) organic resin waste from EPICOR-II
- Highly loaded (>17 Ci/L) inorganic zeolites from the Submerged Demineralizer System (SDS)
- Makeup and Purification System filters and resins totaling >900 nCi/g of transuranic (TRU) material
- TRU and other selected waste materials greater than the limits set by 10CFR61.

In July 1983, shipment of the 50 EPICOR-II waste containers to the Idaho National Engineering Laboratory (INEL) was completed. Disposition of the vessels in high-integrity containers (HICs) is planned to begin in 1984. In August 1983, the 13th SDS waste container was shipped to the DOE site in Richland, Washington, which marked completion of the removal of about 95 percent of the accident generated waste from TMI. During 1983, the purification demineralizer resins were sampled and a system for cesium elution processing, sluicing, and resin removal was designed and built. Operation of the system and resin removal is scheduled for 1984. The DOE is developing a program for characterization of and reimbursable storage and disposal of selected special wastes.

EPICOR-II AND SDS WASTES

Fifty EPICOR-II liners containing 1400 L of organic resin were loaded to a maximum level of 1800 Ci (predominantly Cs and Sr) in the course of processing over 1,900,000 L of contaminated water from the Fuel Handling and Auxiliary Buildings. As a result of radiolysis of residual water and resin degradation, combustible gases (i.e., hydrogen and organic gases) were generated during storage. Methods and equipment were developed and used to remotely purge and inert the stored containers so that all shipments met federal safety regulations. Gas samples were taken to verify that the shipments met safety requirements. The shipments to the INEL were completed in July 1983. The vessels are presently being stored at the INEL prior to their planned disposition in the HICs. The HIC was designed, fabricated, and tested so that the EPICOR-II containers could be safely disposed of in a commercial burial facility as Class C waste as defined by 10CFR61. The DOE is planning a demonstration burial of an EPICOR liner in a HIC at the U.S. Ecology commercial burial facility at Richland, Washington. The EPICOR liner is to be loaded at the INEL and transported in a shipping cask to Richland, Washington. In this plan the remaining liners will be disposed of in a similar fashion by GPU Nuclear.

The HIC is a 2.1-m-high by 1.6-m-diameter reinforced concrete overpack with an internal steel jacket. It is equipped with a hydrophobic gas ventilation system with positive pressure which is installed in the lid. The HIC is designed to be an effective barrier against intrusion and provides for waste stability for 300 years.

In addition a small number of EPICOR-II vessels will be used in a solidification R&D program which is being performed at the INEL to investigate various techniques of immobilizing and stabilizing organic resin wastes. Two vessels have been sluiced and examined for EPICOR container corrosion; no corrosion has been found. Metal tabs have been cut from the vessel and will be examined periodically for degradation from exposure to the resin waste.

Approximately 2,250,000 L of contaminated water from the Reactor Building sump and greater than 1,900,000 L of Reactor Coolant System water were processed through inorganic ion exchangers called zeolites. Nineteen 280-L zeolite containers called SDS liners loaded with radioactive Cs and Sr to a maximum level of 55,000 Ci or to 112,000 Ci if daughter products were included. Because of the radiolytic reaction on residual water, up to 750 cc

(stp) per hour of hydrogen and oxygen gas was generated per container. Equipment and techniques were developed and used for vacuum drying and catalytic recombiner addition. Palladium-platinum catalysts were added to each liner to recombine hydrogen and oxygen and to limit combustible gases to acceptable concentrations thereby ensuring safe conditions for transportation and storage. Gas samples have been taken to verify safe conditions of the liner prior to and after shipment. To date, 14 of these containers have been shipped to Richland, Washington and 3 have been vitrified successfully by Battelle Pacific Northwest Laboratory using an in-can melter. This vitrification was part of a program for immobilizing high-activity wastes. Leach testing of core bores from these glass logs indicated that the leach rates were comparable with defense high-level waste glass logs. In addition to inserting a catalyst, a rupture disc with exhaust filter has been added to each of the remaining 11 SDS liners to ensure safe burial. A pressure transducer has been added to the liner containing the highest curie content, and will be monitored periodically. Also, thermocouples were inserted in the concrete overpack. These 11 liners have been disposed of in a monitored burial facility at Rockwell Hanford Operations, Washington.

MAKEUP AND PURIFICATION SYSTEM FILTERS AND RESINS

During the TMI-2 accident, the Makeup and Purification System filters and demineralizers in TMI-2 were highly contaminated by the letdown of reactor coolant through the system. Quantities of TRU and fuel debris have been found in them. The resins have been surveyed, measured, and sampled while the filters have been removed and examined. A resin sample from the B demineralizer has been examined. GPUNC will remove and DOE will take possession of the purification system resins for reimbursable storage or disposal. In the GPUNC characterization program, a mobile robot called SISI (Surveillance and Inservice Inspection Robot) provided by Westinghouse Hanford Company (WHC) was used to photograph the interior of the demineralizer cubicles, and to take radiation surveys and contamination swipes to determine the room conditions. Prototype non-destructive assay equipment was used to estimate fuel content.

WHC has designed a system to chemically elute the high Cs activity (>21,000 μCi/g of resins), off the resins. The eluate will be processed through the SDS and EPICOR system, the resins sluiced, and the material packaged for final disposition.

A sample of resin and liquid from the TMI-2 Makeup and Purification System B demineralizer vessel was obtained by GPU Nuclear in May 1983 and shipped to Oak Ridge National Laboratory (ORNL) for characterization and experimental studies. Characterization included examining the resins to determine sluicability and chemical composition. The experimental studies included experiments to determine the feasibility of removing Cs from the resins, the compatability of the resulting solutions with the SDS system, the effectiveness of charcoal for removing organics from the solution, and the effectiveness of removing particulates using a sintered stainless-steel filter with a pore size of 0.5 microns.

Batch equilibrium experiments were performed using H_2O, NaH_2BO_3-H_3BO_3 solution, and various strengths of NaOH solution. Both Cs-137 and carbon leaching results were obtained for tests using 1/2 g samples. The first stage, using 20 mL of water, removed some Cs but a second stage was not effective. The NaH_2BO_3-H_3BO_3 solution was 0.035-M NaH_2BO and 0.35-M H_3BO_3 with 800-ppm Na and 3800 ppm B at a pH of 7.6. The first stage removed 53.7 percent of the Cs in the sample and the second stage removed 63.9 percent of the remaining Cs left in the sample after the first stage. Carbon leaching from the first stage was 75 ppm and decreased to 38 ppm for the second stage. The NaOH solutions were more effective than the other solutions as had been expected and the stronger the NaOH solution the better the Cs removal. Carbon leaching also increased with increasing molar concentration of NaOH. Sr-90 removal was much more difficult using the same solutions.

Successive batch stripping of Cs-137 from a 7.5-g sample in a small ion exchange column was performed to simulate an agitated bed process by shaking the column with a remote manipulator. Each stage had 15 mL of strip solution and lasted 1/2 h. Instantaneous stripping decreased from approximately 11 percent for the 1st stage down to less than 3 percent by the 14th stage using sodium borate solution. Cumulative removal by this stage had removed over 50 percent of the Cs. The effectiveness of the 1-M NaOH was then tested and instantaneous stripping increased to over 10 percent for 3 stages before falling rapidly in two final stages which brought the cumulative removal to 93 percent.

Studies by ORNL showed that the removal of carbonaceous materials from the solution by charcoal proved to be ineffective for the requirements at TMI. However, the effects of the organics on the zeolites of the SDS system were minor. The 0.5 micron filter proved to be effective in removing fines from the solution. From approximately 5 mL of 0.5 mCi/mL solution generated from the first 6 stages of the flowsheet tests, a maximum of 1 mg of material consisting of sulfur, titanium, stainless steel, and zirconium was left on the filter. Therefore, a 0.5 micron stainless steel filter will be used to remove material fines and a charcoal filter will not be used in the purification demineralizer Cs elution process.

The Cs elution studies by ORNL showed that sodium borate solutions could effectively lower the Cs concentrations in the resin and therefore make resin removal less man-rem intensive.

As a result, Cs will be eluted from the demineralizers using an eductor (initially used to dilute Cs concentration) and pump (a skid for each vessel) in upflow manner using the sodium borate solution which will be filtered through a 0.5-micron sintered stainless steel filter (back-flushable to remove fuel) and processed through the SDS system. A mobile robot (LOUIE) will be provided by WHC to monitor the gamma activity as the Cs is removed. The filter will be backflushed to remove fuel particles back into the vessels. The remaining TRU contaminated resins will be sluiced in a downflow manner to the spent resin storage tank using existing equipment and packaged for shipment. WHC is providing the design and the equipment for elution. GPU Nuclear will provide for installation and operation of the system and removal of the resin.

SPECIAL TRANSURANIC WASTE MATERIAL

The TRU waste at TMI-2 is different from most TRU waste found at commercial reactor fuel cycle facilities in that it has significant fission product contamination (mostly cesium and strontium) with associated high gamma ray background and contains quantities of boron in the waste matrix. Thus the

DOE will assist in the characterization of TMI-2 TRU waste material and provide for reimbursable shipment, storage, and disposition at the INEL. To date, assistance has been provided in TRU analysis of the Makeup and Purification System filters and resins, the Reactor Building sump and drain tank solids. Removal of 5 makeup filters to the INEL for research and development has been performed. In the near future, the INEL will provide requirements and costs for receipt of the Purification System resins, SDS Cuno filters, and a 6th makeup filter.

CONCLUSION

The DOE research and development program at TMI-2 has involved the processing, examination, packaging, shipment, and disposition of Class C low-level waste and special waste. Processing has included resin elution and sluicing design, and vitrification. Examination has included TRU measurement, gas analysis, radionuclide analysis, and glass leach testing. Packaging has involved the HIC development work and preparations for safe shipment and burial. Shipment and disposition plans have been prepared and are being executed for EPICOR-II and SDS wastes. Retrievable TRU storage will be provided on a reimbursable basis for the special TRU wastes.