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EMERGENCY ACTIONS CONCERNED WITH EFFLUENT CONTROL AT TMI

R. E. Brooksbank, D. O. Campbell, J. W. Snider, O. O. Yarbro

Chemical Technology Division
Oak Ridge National Laboratory*
Post Office Box X
Oak Ridge, Tennessee 37830

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Introduction

Shortly after the TMI accident, it was recognized that the status and objective of the TMI site was transformed from that of being an electrical power-producing function to that of being a radiochemical processing plant. The rupture of the reactor's fuel cladding within the reactor vessel and the subsequent transfer of fission products to the primary coolant as the accident proceeded brought about this change. As the accident continued, the contaminated water also found its way into both the Reactor Containment Building and the Auxiliary Building. In addition to the problems brought about by contaminated water, volatile gaseous fission products (I, Xe, and Kr) were released from the reactor system, which represented a potential problem for the environment.

On March 30, 1979, 2 days after the accident, requests for chemical engineering assistance to TMI were formally made by several concerned organizations, including General Public Utilities Corporation (GPU), NRC, DOE, and the Electric Power Research Institute (EPRI). Because many of the problems created by the accident pertained to radiochemical engineering, the Chemical Technology Division[1] of the Oak Ridge National Laboratory was selected to provide this service. The first chemical engineers were sent to the site on April 1, 1979. As work continued during the emergency

[1] R. E. Brooksbank and L. J. King, Involvement of the ORNL Chemical Technology Division in Contaminated Air and Water Handling at The Three Mile Island Nuclear Power Station, ORNL/TM-7044, (August 1979).

and other tasks of a chemical engineering nature were recognized, additional staff members were requested for on-site assistance. In addition to the on-site assignments, other chemical engineering staff members provided backup support to these individuals. Personnel from the Analytical Chemistry Division and senior flowsheet chemists were also employed on a continuous basis at ORNL to interpret the data obtained from the many samples sent to ORNL from TMI. The major objective of the on-site chemical engineering group was to provide advice and guidance to the technical management staff of GPU. Efforts to the waste management group were specifically aimed at effluent control problems. Their objectives included

1. stopping the release of ^{131}I to the environment;
2. providing conceptual designs for water storage and processing systems that were adequately contained;
3. providing guidance for handling surface and equipment contamination.

ORNL assistance to TMI continues within the chemical engineering areas of flowsheet development and verification of processes prepared for use in the decontamination of high-activity-level water (HALW).

As the result of this experience, a series of recommendations have been made to improve the operations within the nuclear option as they relate to potential emergencies. The Department of Energy is actively reviewing the "lesson learned" from the accident and

the ongoing post-accident cleanup experience[2] in an attempt to establish generic needs for improved systems. A discussion of the technical conditions that existed after the accident coupled with an understanding of the various tasks undertaken as they relate to effluent control might serve as a meaningful exercise.

Summary of Critical Emergency Requirements

In looking back at the hectic days following the accident, two major categories emerge as being significant from an emergency standpoint, namely (1) the requirement to stabilize the reactor system, and (2) to minimize the radiological impact on the environment. Figure 1 presents a breakdown in summary fashion of these critical requirements.

Reactor stabilization.[3] – Two days after the accident, the reactor fuel temperature was 360°F with the only heat source being the fission product decay process. The reactor primary coolant circuit was operating at a temperature of 280°F and the pressure was being maintained at 100 psig. Limited amount of gaseous radioactivity was being released to the atmosphere from two sources in the Auxiliary Building. The first release was from the liquid transferred during the early phases of the accident.

Additional equipment was installed on a crash basis to provide long-term cooling and pressure control to the crippled reactor systems.

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[2] R. E. Brooksbank and W. J. Armento, Post-Accident Cleanup of Radioactivity at The Three Mile Island Power Station, ORNL/TM-7081, (February 1980).

[3] F. J. Patti, A. S. Dam, and E. C. Brolin, "Securing Three Mile Island: the initial recovery programme," Nuclear Engineering International, (November 1979).

Cold shutdown is normally attained by using the decay heat removal system. In the TMI case, it would normally start operating once the reactor coolant pressure decreased to 300 psig. Since a significant amount of piping and equipment in this system is located in the Auxiliary Building, its use under post-accident conditions might have unnecessarily contaminated the system and increased personnel radiation dose rates in the Auxiliary Building. Since this would have limited and made work in this building more difficult, it was decided that the decay heat removal system should not be used unless absolutely necessary.

Accordingly, back-up systems were designed and installed for both temperature and pressure control under emergency post-accident conditions.

Because this paper is concerned about the latter category on this figure, the remainder of this discussion will relate to the engineering aspects of the control of both gaseous and liquid effluents from the site.

Gaseous Effluent Status and Control

The control of radioactive off-gases has been a primary concern in the field of radiochemical reprocessing of irradiated fuel, and chemical engineers with this experience background were intially assigned the task of assisting in this vital area.

An assessment of the condition of the off-gas handling and treatment system and support buildings was begun shortly after the accident and is still in progress. The immediate problem following

the accident was the release of iodine and the noble gases in excess of release specifications for normal operations. Because iodine has a more pronounced effect on the health and welfare of the downstream population, serious attention was given to the effectiveness of the charcoal traps designed to remove this isotope. Both downstream and upstream samples of the charcoal traps contained in the Unit 2 Auxiliary 2 and Fuel Handling Buildings, through which all gaseous releases from TMI-2 emanated, indicated that the traps were ineffective in removing the iodine. Problems inherent in establishing the effectiveness of the off-gas removal systems involved high radiation levels surrounding both the monitoring equipment and the traps themselves. Figure 2 shows a schematic representation of the off-gas system immediately following the accident.

Results of the tests conducted on the iodine trapping efficiency of the charcoal units within the Unit 2 Auxiliary and Fuel Handling Buildings indicated that all the traps should be replaced. Therefore, a total of 300 traps were changed (180 in the Auxiliary Building and 120 in the Fuel Handling Building) throughout the period April 20 to May 3.

Because the reactor system had not yet stabilized from the standpoint of the natural convection cooling mode and the primary loop contained an estimated 6.5×10^7 Ci of iodine, the decision was made to provide the existing off-gas trains with a supplemental system. This system, which contained four trains with a total treatment capacity of 100,000 cfm, was located on the west coast and was transported to the site from Pasco, Washington. It was flown to the TMI-2 site for installation on the Unit 1 Auxiliary Building roof and was placed on-stream on May 3, 1979. Currently, three of the four

trains are in operation. Figure 3 summarizes, in schematic fashion, the overall modifications made to the off-gas system. An additional modification (also shown in the figure) is the capping of the stack vent; this provided an extra margin of safety. Figure 4 is a photograph of the supplemental filter installed on Auxiliary Building roof. Figure 5 represents, in schematic fashion, a history of the ^{131}I releases and the results of the modifications.

From the previous figure, it is clear that the iodine release ended when the new supplemental filters were placed on-stream. Before that, only about 15 Ci was released, a relatively small amount. The total actual release of major radioisotopes is shown in Fig. 6. The fission product core inventory, based on ORNL ORIGEN calculations is shown in the first column of figures. The second column is the quantity that has actually been accounted for as measured in the water, the Containment Building atmosphere, or released from the plant. About 70% of the tritium and noble gases was released from the fuel and 60% of the cesium. We believe that the iodine must have been released to a comparable extent, but only 40% has been accounted for; that is, dissolved in the water. We suspect that the missing 20-30% precipitated, probably as silver iodide, and therefore has not been sampled. Early into the accident, sodium thiosulphiae-sodium hydroxide solution was put into the Auxiliary Building sump and on the floors as an emergency action to render the iodine in solution non-volatile. We have not fully evaluated the effect of this "holding" reagent in the TMI situation.

Finally, in the right hand column of this figure the amount that escaped the plant is shown. About 8% of the Xe was discharged to the atmosphere, but only 10^{-5}% of the iodine. Clearly, iodine was held up inside the system to a very large extent, and this is because of its chemical interaction with water to give a non-volatile species.

As an aside, it may be significant to compare the ^{85}Kr now in the Containment Building atmosphere (~57,000 Ci), which is now the subject of much controversy.

As part of our technical assistance to the Kemeny Commission, the path of iodine under TMI's conditions and in the event of a reactor "meltdown" were briefly studied. The iodine release pathways (Fig. 7) must be considered to understand the environmental effects. Fission products were released from the fuel into a very hot-gas phase consisting of steam and hydrogen. The gas was then cooled as it mixed with the primary coolant water. A mixture of primary coolant and gas went into the Containment Building, through the infamous PORV that had stuck open and a rupture disc; this material is still in this location.

Some water was transferred from the Containment Building to the Auxiliary Building, but this was not the major contributor. The largest source was probably by way of the primary water letdown and makeup system, along with leakage. This permitted release into the Auxiliary Building atmosphere of the noble gases dissolved in the water, along with a small fraction of the iodine that volatilized from the water. As stated previously, the effect of the iodine holding chemicals

added to building floors and sump were not evaluated, but past experience would indicate that some benefit

Basically, the release was from the fuel to the hot gas to water to air and the air was exhausted through ineffective charcoal filters. The small iodine release was a result of the stability of the non-volatile species in the water. In contrast, noble gases were not held up by water or charcoal, and they were

A recent report by Stratton[4] concludes that in the event of a meltdown:

> "If a large amount of alkaline water is present and if the Containment Building maintains its integrity, the iodine release will be very small regardless of the damage to the reactor core and primary system because:
>
> 1. The major iodine release path was via the primary letdown over many days, but letdown would cease if the core vessel system were penetrated.
> 2. Silver in the control rods would vaporize, disperse in the system. Silver would then react with the iodine, forming immobile gI. The analysis of the water presently in the containment vessel <u>does</u> indicate the presence of control rod constituents (Ag, In, Cd).
> 3. Much greater use of the Containment Building spray system, as in the case of a meltdown, would decrease iodine volatility by a large factor ($\rightarrow 10^3$)."

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[4] W. R. Stratton, N. H. Fontana, R. L. Seale, A. B. Reynolds, "Alternative Event Sequences or What More Could Have Gone Wrong," American Nuclear Society-European Nuclear Society Topical Meeting on Thermal Reactor Safety, Knoxville, TN., April 7-11, 1980.

Liquid Effluent Status and Control

As is well known, large volumes of contaminated water were produced from the TMI accident, largely as the result of the release of fission products to the reactor primary loop and the subsequent release of this liquid to the containment and the auxiliary buildings. The problem of water handling[4] was one of primary concern during the early critical stages of the emergency; however, flowsheet development for processes to be used for the decontamination of this water still remains an item of concern 1 year following the incident. Again, as was the case in the treatment of off-gases, some background relative to the status of the water at the time of the accident is necessary in order to fully appreciate the emergency actions taken.

Contaminated water was continually being generated at TMI following the accident because of leakage through pump seals, flushing of sampling systems, and flushing of contaminated floor areas. The major concern relative to this water was that the quantity which could be accumulated might exceed the storage capacity. There was also concern that the water level in the Containment Building might rise high enough to render some vital instruments inoperative. The eventual need to treat all the liquids, including the primary coolant and all decontamination solutions, was considered throughout the planning for water handling.

The status of the liquid handling systems as of April 1, 1979, is shown in Fig. 8. The locations of pertinent areas are designated

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[5]R. E. Brooksbank, "A Special Radioelement Problem: ORNL Assistance to Three Mile Island in Handling Contaminated Air and Water," presented at The Twenty-Third Conference on Analytical Chemistry in Energy Technology, Gatlinburg, TN, Oct. 9, 1979.

in Fig. 9. The primary reactor coolant loop contained 87,000 gal of highly radioactive coolant with an ^{131}I inventory of $\sim 6.5 \times 10^7$ Ci. In addition, the Reactor Containment Building was estimated to contain ~225,000 gal of water which had been contaminated by a large volume of the radioactive reactor coolant. Some instruments were inoperative, probably because they were submerged. The tanks in the Unit 2 Auxiliary Building were full, and floor areas had become flooded with water that had either overflowed or leaked from the tanks. Portions of this water were contaminated to varying degrees by water that had been transferred from the Containment Building sump during post-accident operations before the Containment Building was isolated. The Unit 2 Reactor Building went into containment ~4 hr after the accident and has remained in this state ever since.

The Unit 1 Reactor, which had been shut down for refueling prior to the accident, was being brought up to operating temperature by the reactor coolant pump energy input prior to going critical. The available tankage within the Unit 1 Auxiliary Building was becoming filled with water due to normal operation.

None of the Unit 2 water could be treated. The Unit 2 reactor coolant letdown stream could not be treated because of mechanical problems in the Unit 2 reactor coolant letdown evaporator. The other liquid wastes originating in Unit 2 are normally treated in the Unit 1 miscellaneous waste evaporat; this evaporator, however, was out of service because a demineralizer bed was being changed. In any case, the transfer of Unit 2 post-event water to the Unit 1 Auxiliary Building was considered to be undesirable.

Water inventories in both auxiliary buildings were increasing. There was an urgent need for additional storage and/or water treatment facilities. In accordance with this requirement, an emergency tank farm system was designed and installed in an unused fuel storage basin to provide an additional 110,000 gallons of storage capacity. Figure 10 presents an equipment schematic and Fig. 11 is a photograph of the lower tanks during installation.

In order to proceed with the recovery of the reactor system, the liquid being held in the Containment Building will require removal and treatment. Currently, this liquid is standing at a depth of approximately 7-1/2 to 8 ft and is covering several components, including instruments. Although the leakage of water into the containment area has been minimized, the possibility for increased leakage continues to exist. In the early phases of the accident, the water in containment had ^{131}I concentrations estimated to be of the order of 10^4 μCi/ml. Because of this factor, every effort was made to avoid disturbing this solution until the radioiodine had been allowed to decay. Figure 12 shows the fission product decay curves, based on the analysis of primary coolant, for the radioactive nuclides that were of greatest concern with regard to treatment of the solution and indicated the most desirable processing period.

Water Treatment Processes

The treatment of low-level activity water (10^{-4} μCi/cc ^{137}Cs) has proceeded without difficulty and without impact to the environment.

The treatment of intermediate activity level water (<100 μCi/cc ^{137}Cs) to date has resulted in a liquid process effluent that is below technical specification limits, however, none of this liquid has been discharged because of political and legal concerns. The process called Epicor-II employed for treatment of this lqiuid uses a 3-stage system composed of a mixture of cation, anion and inorganic exchangers. Because of the collection and concentration of fission products in this process high radiation fields result and can conceivably cause an impact on the environment. A little used building was converted in accordance with reprocessing plant criteria, to permit the actual processing to take place remotely and to provide adequate containment to prevent environmental insult. The building was sealed to operate at negative pressure with respect to the outside and independent off-gas trains were installed for the treatment of gaseous effluents from the process. The installation of this process and adaption of this facility may be regarded as an emergency action.

A considerable amount of emergency technical effort[6] has been accomplished to prepare for the decontamination of high-activity level water which contains large quantities of the cesium and strontium isotopes. The first of several samples from the reactor coolant system (RCS) were obtained within a few days of the accident but the larger quantity of water in the Containment Building floor could not be

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[6] E. D. Collins, J. E. Bigelow, D. O. Campbell, L. J. King and J. B. Knauer, "Flowsheet Development Studies for The Decontamination of High Activity Level Water at Three Mile Island Unit 2," AIChE, Portland, Oregon, Aug. 17-20, 1980.

sampled until an access probe was installed about five months later. Several samples were sent to ORNL for chemical and radiochemical analysis. A team of chemists and chemical engineers at ORNL, in conjunction with the Technical Advisory Group to the TMI-2 Recovery Staff of GPU initiated potential flowsheets and recommended a clarification-zeolite ion-exchange process. A processing system was designed by Allied General Nuclear Services (AGNS) for Chem-Nuclear Systems, Inc. (CNSI), the prime contractor for fabrication, installation and operation of the process equipment. The processing facility, which is called the Submerged Demineralizer System (SDS), is being installed in one of the spent-fuel storage pools at TMI.

Basically, the technical flowsheet work done for the conceived SDS flowsheet followed the information contained on Fig. 13. Sorbents available for suitable application had to be selected based on the exact characteristics of TMI water, suggested flowsheets were required and ion exchange studies were necessary. Finally, hot-cell ion-exchange tests and filtration tests were necessary to provide verification of the designed system.

The characteristics of the high level water generated as the result of the TMI-accident are unique to the nuclear industry. A photograph of the water removed from the top, middle and bottom of the TMI-acccident are unique to the nuclear industry. A photograph of the water removed from the top, middle and bottom of the Reactor Containment Building is shown on Fig. 14. Both the water in the primary loop and the Containment Building have high levels of sodium borate and boric acid, with a pH of 8. The analysis of the high activity level water contained in this solution is shown on Fig. 15. The solids analysis is shown on the next slide (Fig. 16).

Based on the technical work accomplished, which could easily cover an entire session, the flowsheet was selected and designed. Figure 17 presents this flowsheet. This flowsheet will take the contaminated water and clarify the solution by filtration during transfer with the ion-exchange feed tank. The clarified water will be pumped through either or both of the trains of ion-exchange columns. Each train consists of a series of three columns containing zeolite and a column containing an organic cation exchange resin. Finally, the effluent water from each train is combined and passed through a large polishing column containing layers of cation, anion and mixed resins. The operating procedure provides that 200 bed volumes of water will be passed through each zeolite column while it is in position. At that time, the column containing the loaded zeolite will be removed from the system and the other zeolite columns moved forward one position and a new column installed in the third position. In this manner, the zeolite columns will sorb most of the cesium and those in the second and third positions will allow the necessary residence time for ^{90}Sr sorption.

Conclusions

The basic conclusions which can be drawn from the emergency actions concerned with effluent control at TMI are presented in Fig. 19.

One of the first conclusions to be drawn is that the utility recognized the need for technical assistance early in the post-accident period. Had this not been the case, the severity of the accident would have had a more pronounced effect on the environment. In areas affecting

the health and welfare of the public, the utility management was responsible to the advice and guidance provided by experts and translated this information interms of hardware, regardless of the costs. The installation of the supplemental off-gas system to restain the iodine on the site at a cost approaching five million dollars is an example of this responsibility.

When reviewing the basic emergency actions taken on the site, we believe they were significant in reducing the potential and effective impact on the environment.

Although many problems continue to exist at TMI, we believe that a wealth of technology exists, or can be developed to permit the clean-up of TMI-2.

Finally, we believe that the experience obtained from the accident and the emergency responses taken will do a great deal towards improving the safety of the nuclear option.

SUMMARY OF CRITICAL EMERGENCY REQUIREMENTS

- STABILIZE REACTOR
  - TEMPERATURE CONTROL
  - PRESSURE CONTROL

- RADIOACTIVITY RELEASED TO THE ENVIRONMENT
  - GASEOUS EFFLUENT CONTROL
  - LIQUID EFFLUENT CONTROL
  - SURPLUS WATER MANAGEMENT
  - FACILITY CONTAINMENT

FIGURE 1

ORNL Dwg 79-13229AR1

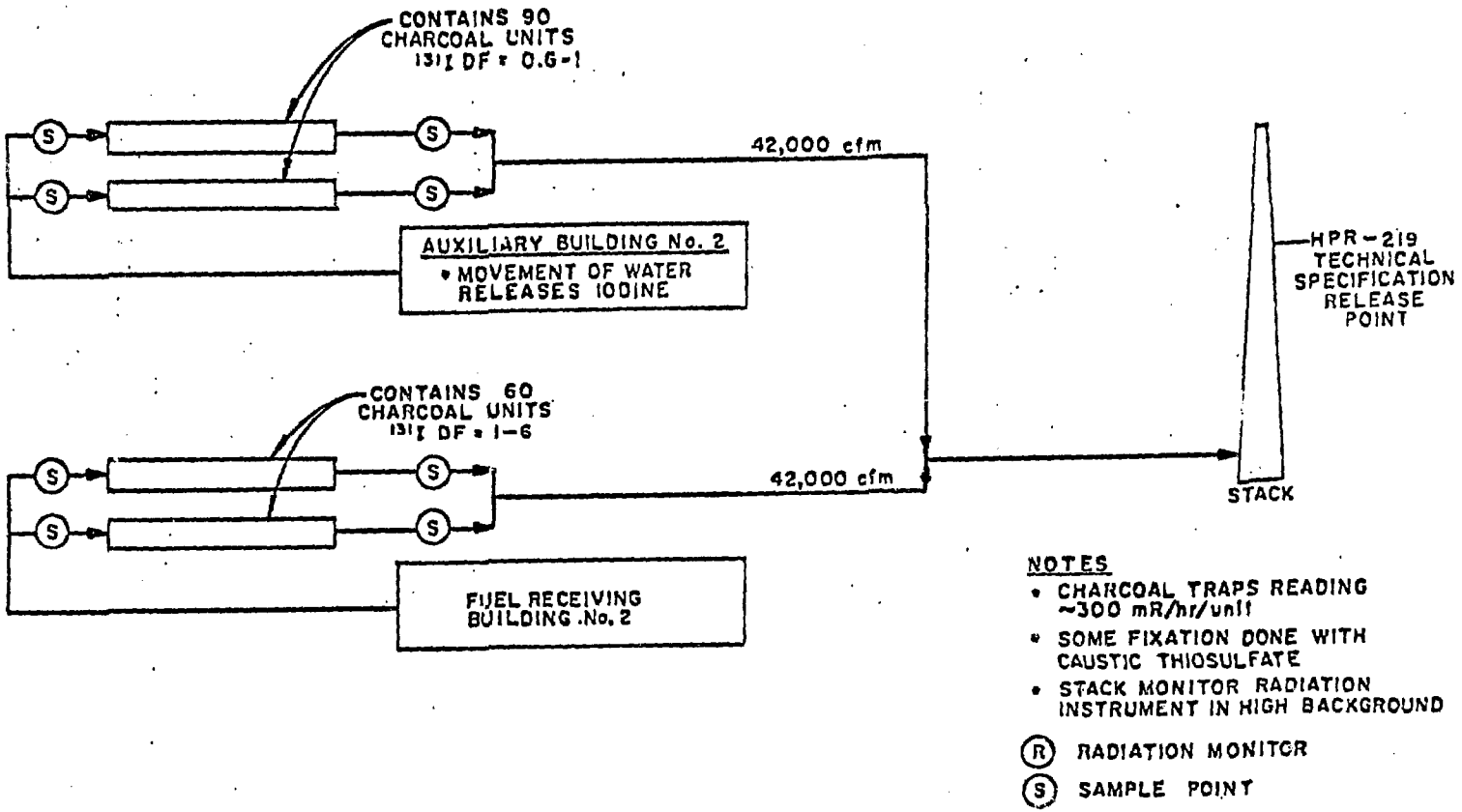


Fig 2. Off-gas system following accident.

ORNL Dwg 79-13230A

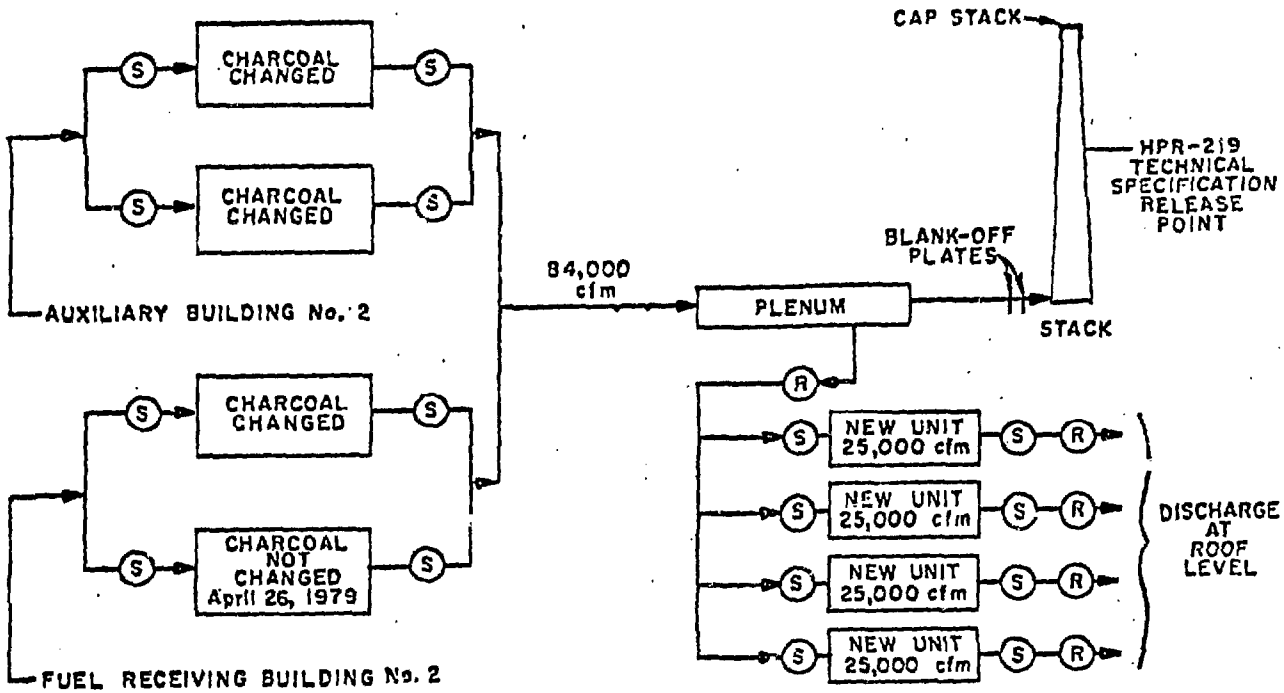
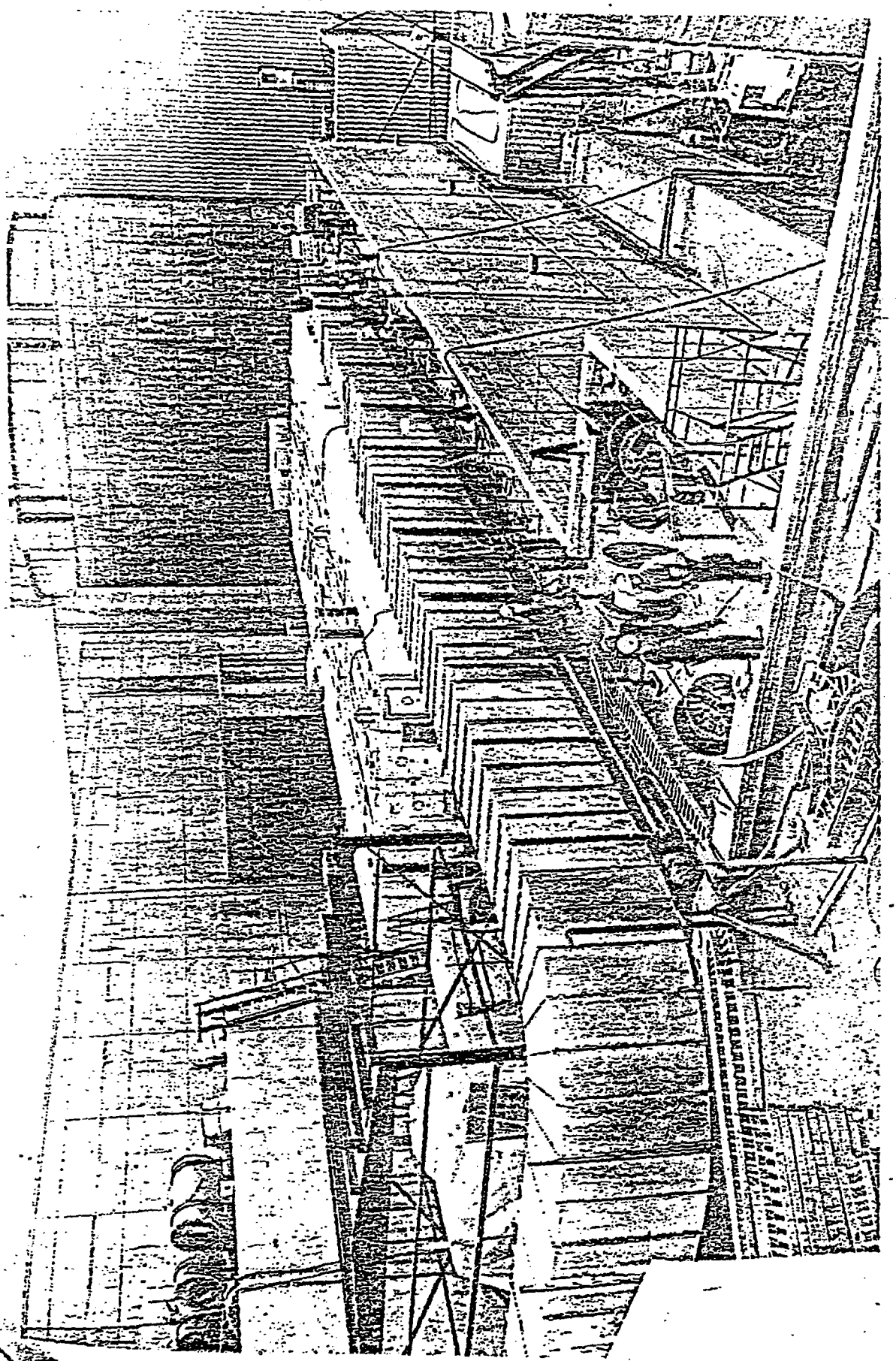


Fig 3

~~Fig 2.~~ Off-gas system modifications.



ORNL Dwg 79-13231R2

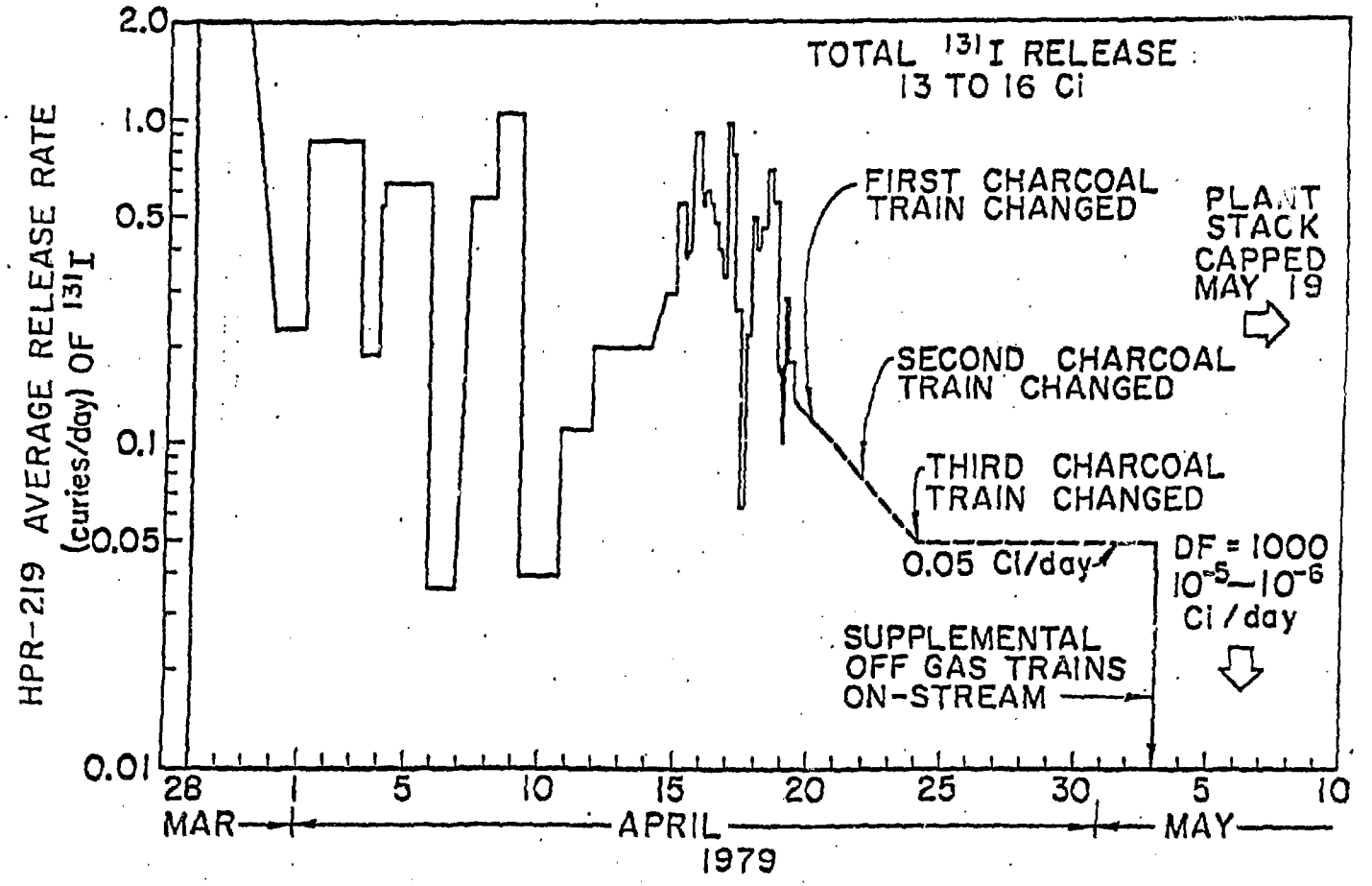


Fig. 3. Release of ^{131}I.

Fig. 45

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ACTUAL RELEASE OF MAJOR ISOTOPES AT TMI

Isotope	Core inventory (Ci)	Measured Activity ~~Released from fuel~~ Ci	%	Escaped to environment Ci	%
^{3}H	~~3750~~ 3.8E2	2740	73	—	—
^{85}Kr	9.6E4	6.8E4	71	—	~8
^{133}Xe	1.5E8 ✓	9.9E7	68	1.2E7*	8.2
^{131}I	6.5E7 ✓	2.7E7	42	15	2.3E-5
^{137}Cs	8.5E5	5.1E5	60	~0	~0

*If released at time of accident, 8.3E6 Ci actually released over a period of time.

Figure 6

ORNL Dwg 79-158890

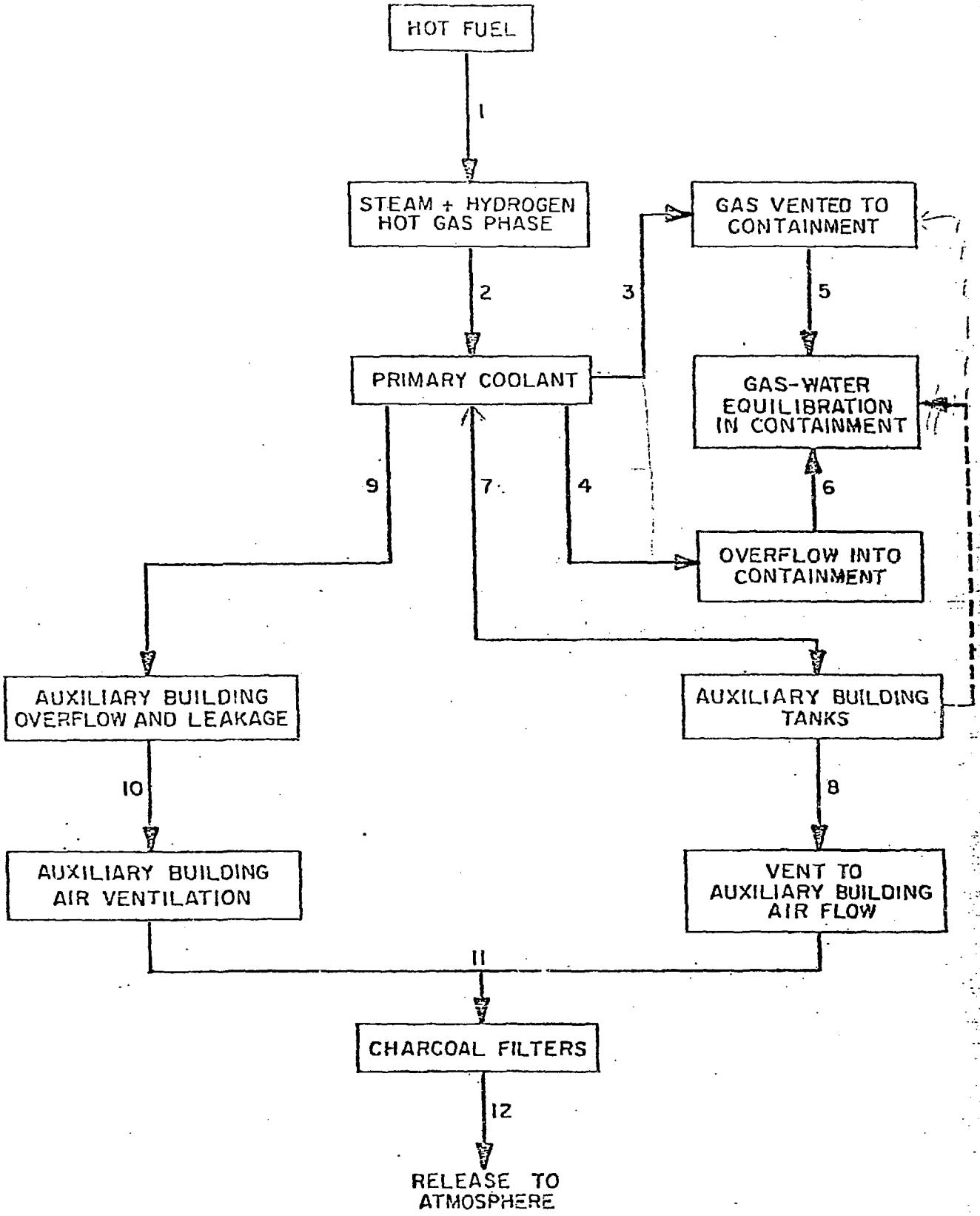


Fig. 7 Iodine transport paths

ORNL Dwg 79-13232AR

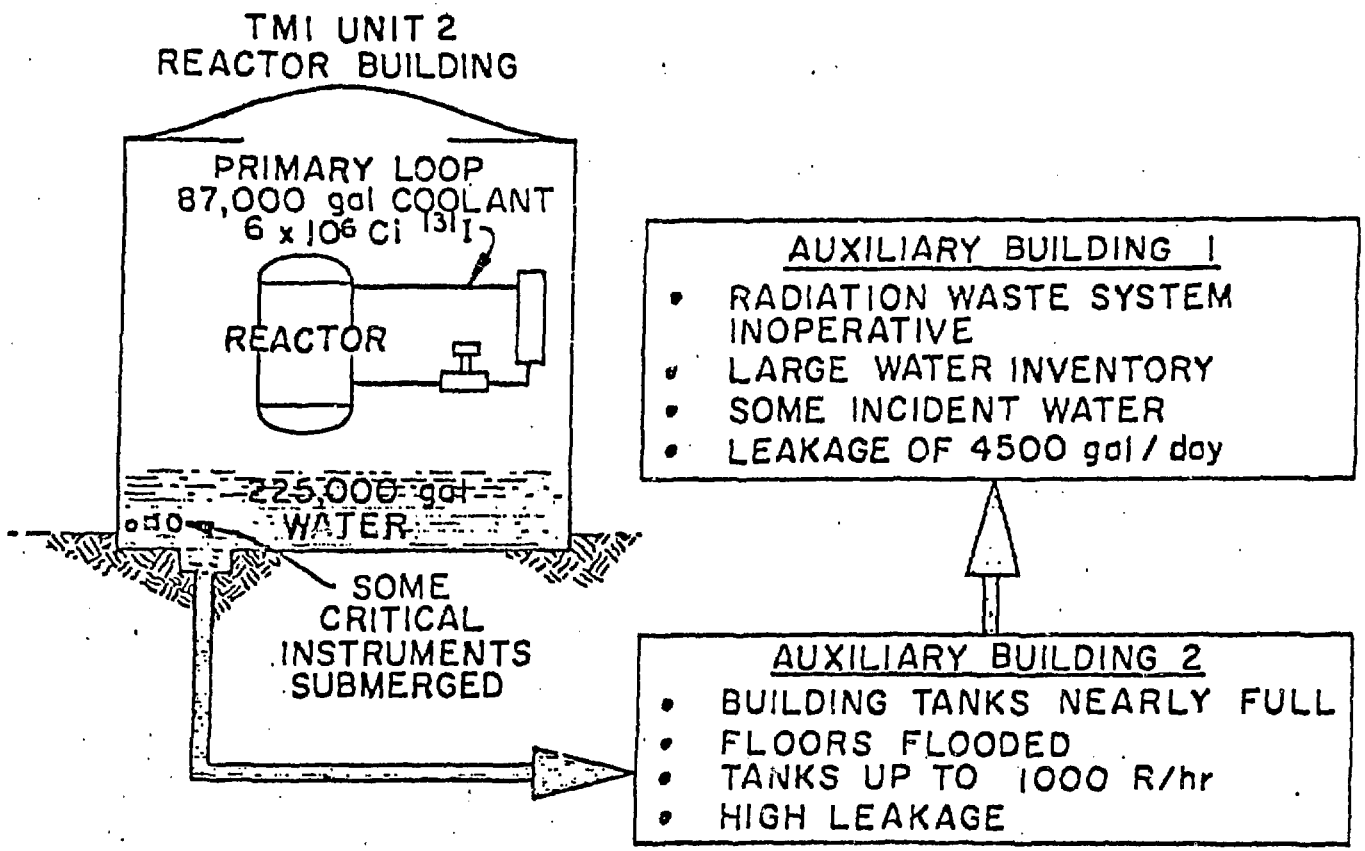


Fig. 8. Status of liquid at TMI following the accident.

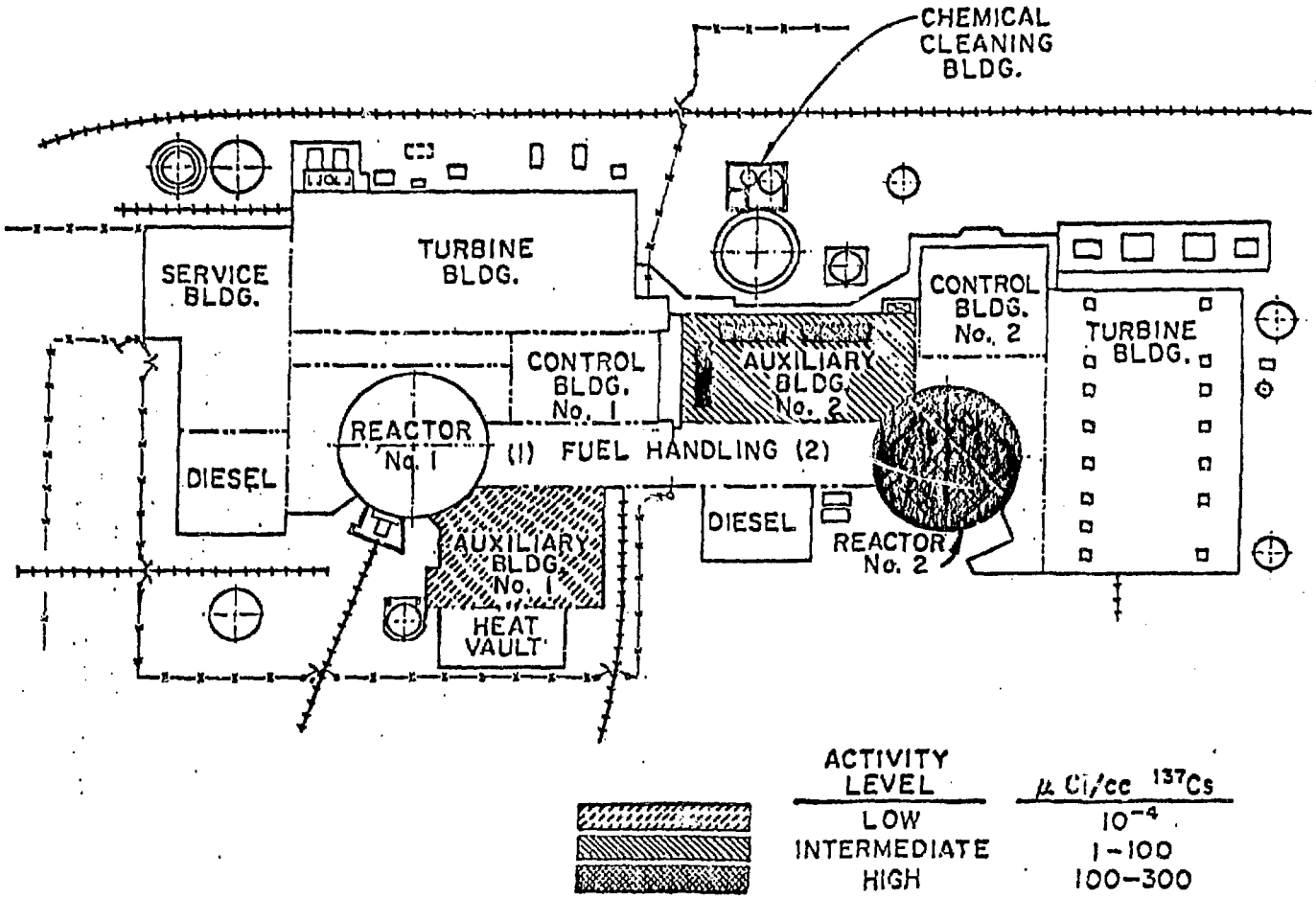


Fig. 9 Distribution of contaminated liquid

ORNL Dwg 79-13241A

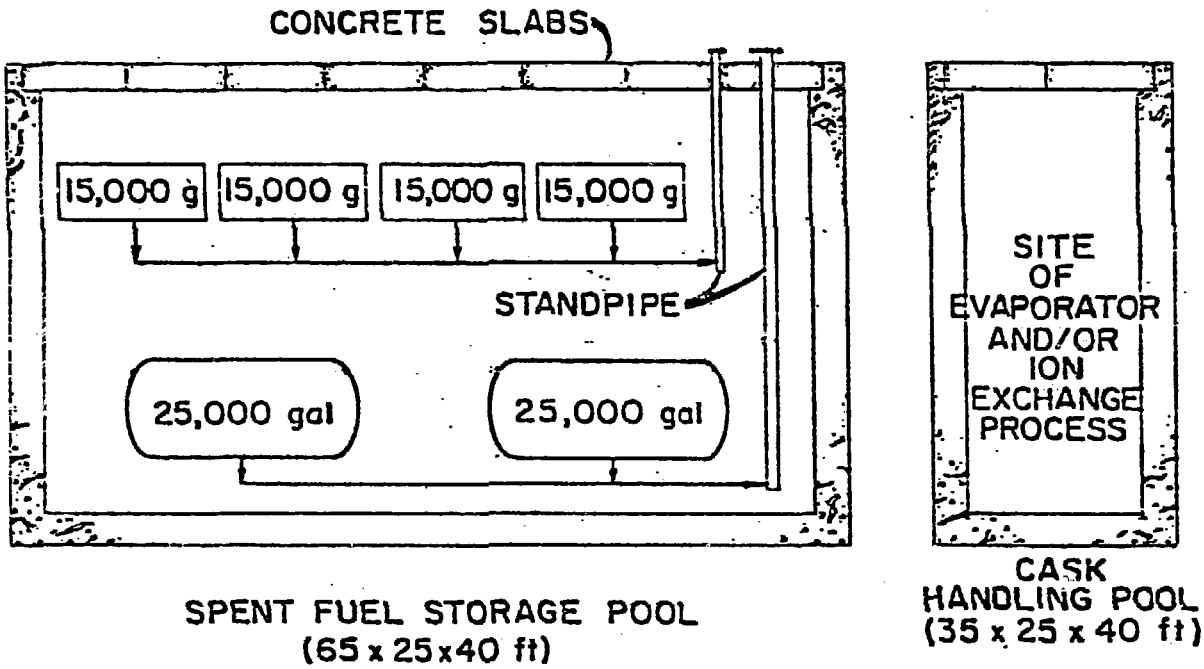


Fig. ~~11.~~ 10 Emergency tank farm at TMI Unit 2.

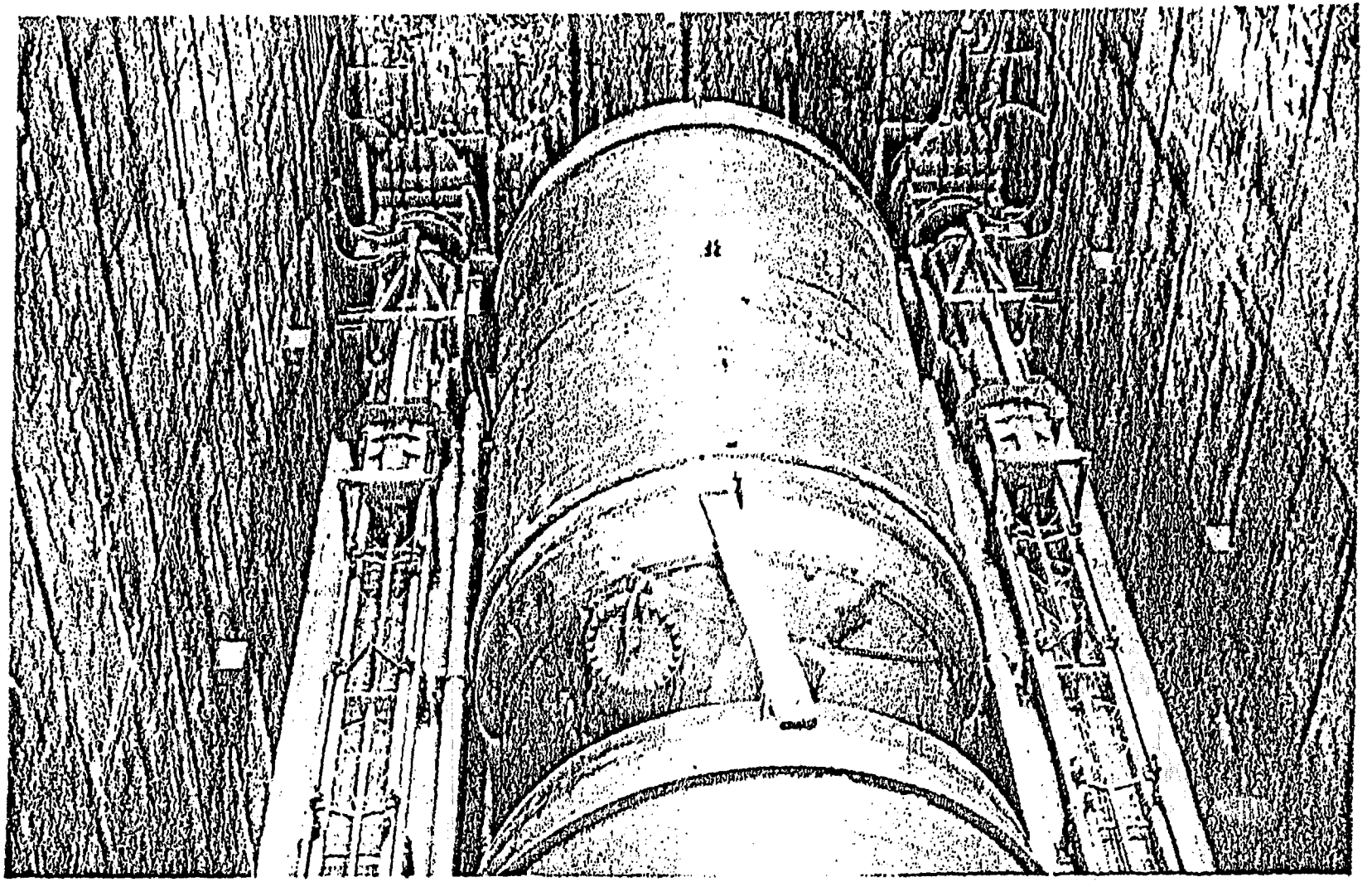


Fig. 11. Tank farm in the TMI Unit 2 fuel storage pool during installation.

ORNL Dwg 79-13239A

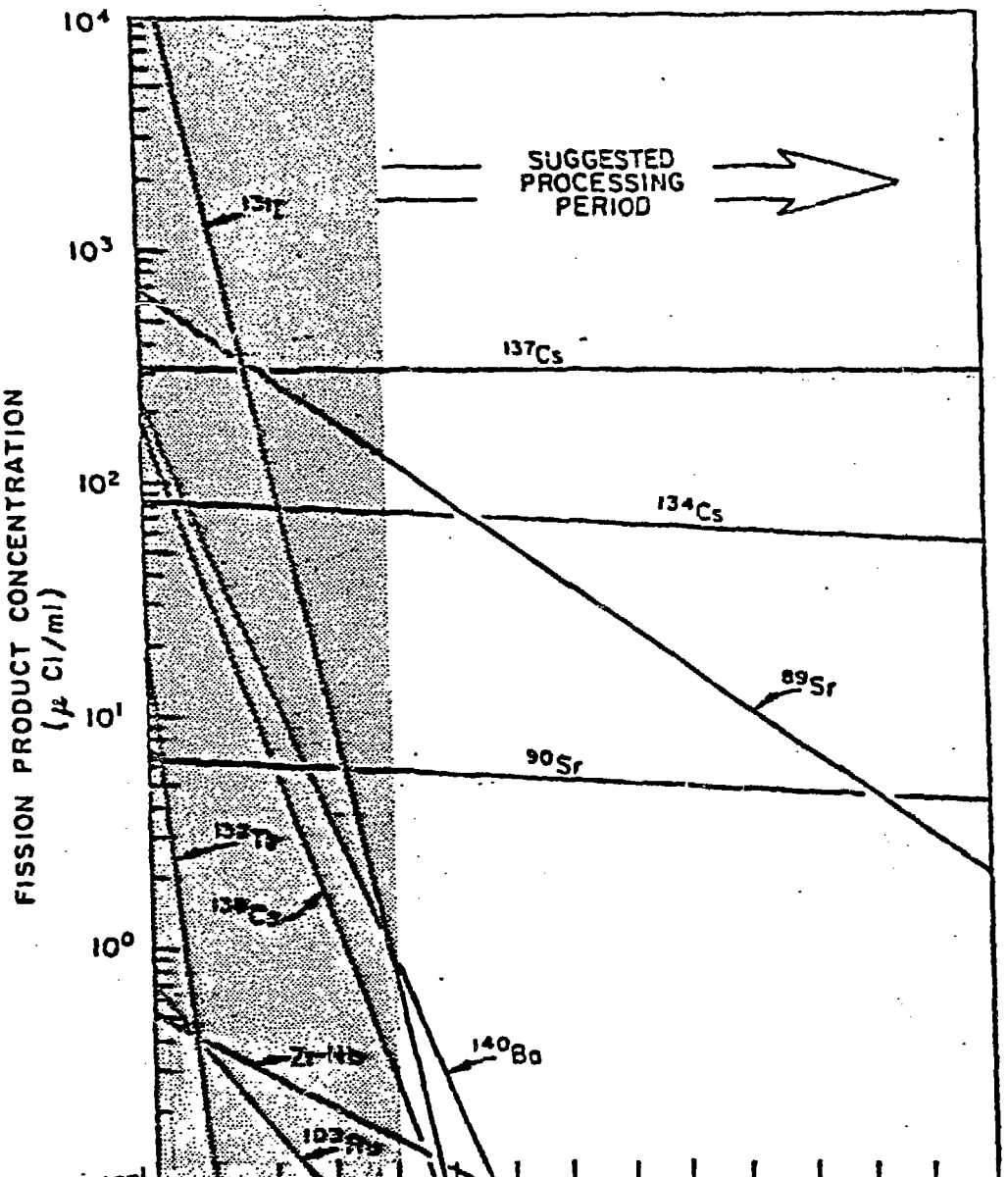


Fig. 12 Fission product decay in the TMI-2 primary loop.

ORNL Dwg 80-12343

PERTINENT HISTORY

DISTRIBUTION COEFFICIENT (K_d) STUDIES

ALTERNATIVE FLOWSHEETS–

TRACER-LEVEL ION EXCHANGE STUDIES

SDS FLOWSHEET VERIFICATION TESTS

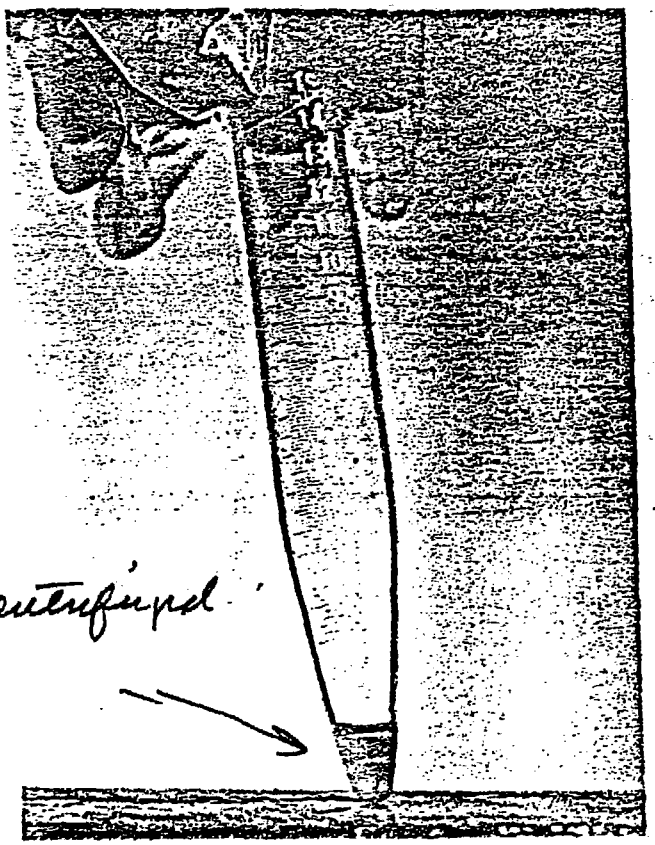
ORNL PHOTO 4320-79

TOP

MIDDLE

Physical appearance.

BOTTOM



CENTRIFUGED SOLIDS

ORNL Dwg 79-15906

LABORATORY MEASUREMENT*

ISOTOPE	TOP SAMPLE	MIDDLE SAMPLE	BOTTOM SAMPLE	PREDICTED VALUES
^{137}Cs	176	179	174	200–260
^{134}Cs	40	40	39.6	30–40
^{140}La	0.09	0.078	0.014	0.25–0.49
^{3}H	1.03	1.05	1.01	1.0–1.5
^{131}I	0.012	0.012	0.013	0.015–0.044
^{90}Sr	2.70	2.90	2.83	10–18
^{89}Sr	43.6	40.6	42.1	171–228

* μCi/ml

Core damage less than expected based on Cs - Sr isotope at pH 8 is unstable

30

ORNL Dwg 79-15899

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NUCLIDES IN BOTTOM SAMPLE PRECIPITATE

(μCi/ml OF TOTAL SAMPLE VOL)

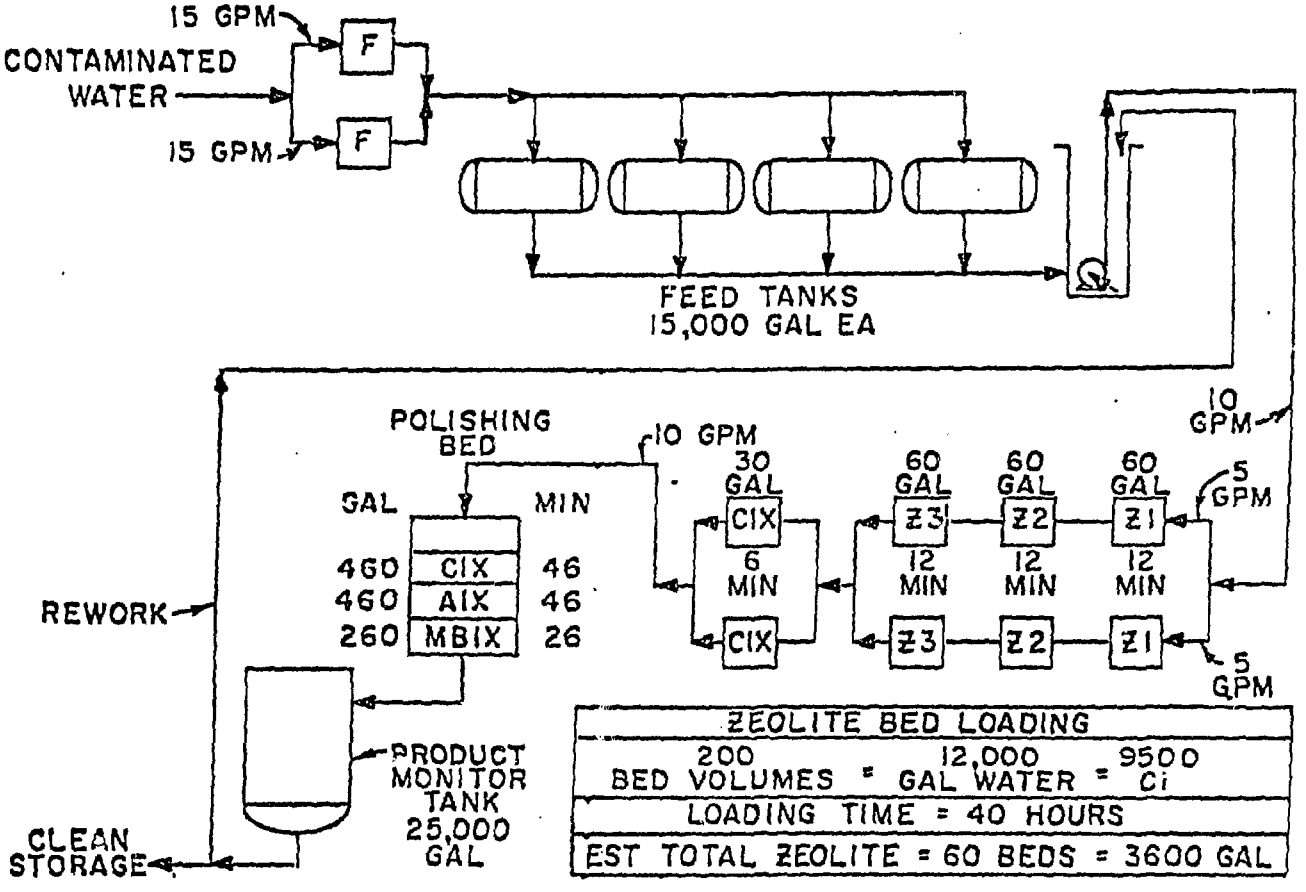
^{95}Zr	0.037/0.061
^{95}Nb	0.104/0.162
^{129m}Te	0.28/0.51
^{140}La	0.11/0.12
$^{89+90}Sr$	2.78

fission products.

(μg/ml OF TOTAL SAMPLE VOL)

In	0.16 ✓ also Ag, Cd control rods.
Cu	54
Mn	0.6
Al	7

Predominant (tailing?)



SDS FLOWSHEET

Conclusion

- The Utility recognized the need for technical assistance early in the post-accident period.

- Emergency actions taken at the site resulted in minimizing the impact on the environment.

- Technology exists or can be developed for an orderly, effective cleanup of the TMI-2 ~~system~~ site without effecting the environs.

- The lessons learned from the accident and the emergency actions taken during the emergency can do much towards enhancing the nuclear option.