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RECENT CHEMICAL ENGINEERING REQUIREMENTS AS THE RESULT OF TMI ON-SITE EXPERIENCE

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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. The major role of the chemical engineering discipline under these conditions then became one of effluent control during these early phases.

As the investigative work concerning the post-accident period comes to light, it will become apparent that chemical engineers and flowsheet and analytical chemists played a significant role in the recovery of TMI. Work in the area of effluent control, waste handling, analytical service, flowsheet development, and decontamination will all be regarded as significant. Successful

efforts to assist TMI in these specific areas were made possible by the close working relationship between chemical engineers and chemists.

Chemical Engineering Assistance Requested as the Result of the Accident

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 and other tasks of a chemical engineering nature were recognized, additional staff members were requested for on-site assistance. A total of ten chemical engineers from this task group participated in the emergency operations over a critical 3-month period. 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 of 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 facilities 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 "lessons learned" from the accident and the ongoing post-accident cleanup experience[2] in an attempt to establish generic needs for an improved system. Many of the topics selected for this session directly resulted from the on-site involvement of chemical engineers at TMI. Perhaps a discussion of

the technical conditions that existed after the accident coupled with an understanding of the various tasks undertaken by chemical engineers might serve as a meaningful exercise toward understanding the future role of chemical engineers.

Control of Gaseous Effluents

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 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 1 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 6×10^6 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 in Pasco, Washington. It was flown to the TMI-2 site for installation on the Unit 2 Auxiliary Building roof and was placed on-stream on May 3, 1979. Currently, three of the four trains are in operation. Figure 2 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 3 represents, in schematic fashion, a history of the ^{131}I releases and the results of the modifications.

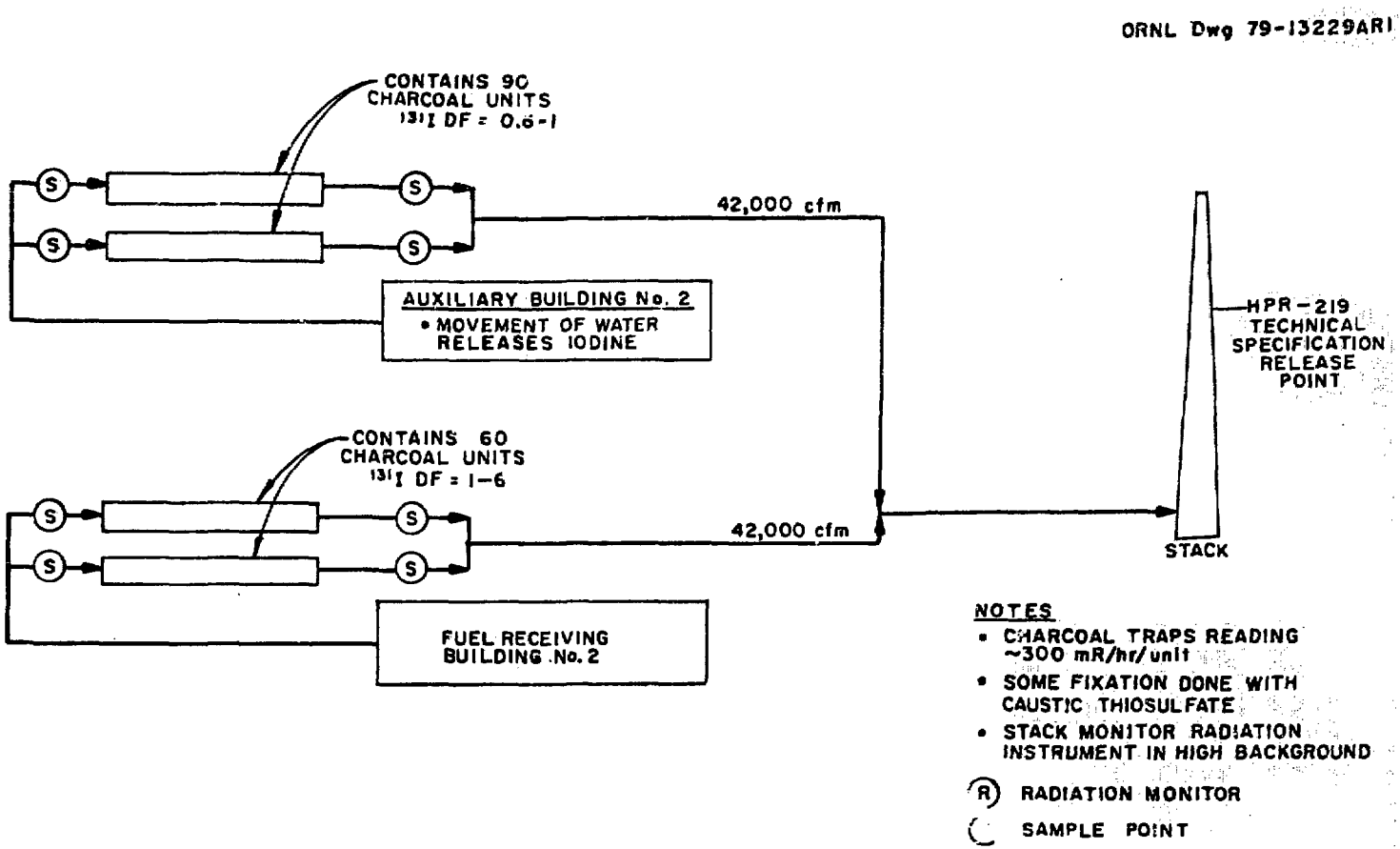

Fig 1. Off-gas system following accident.

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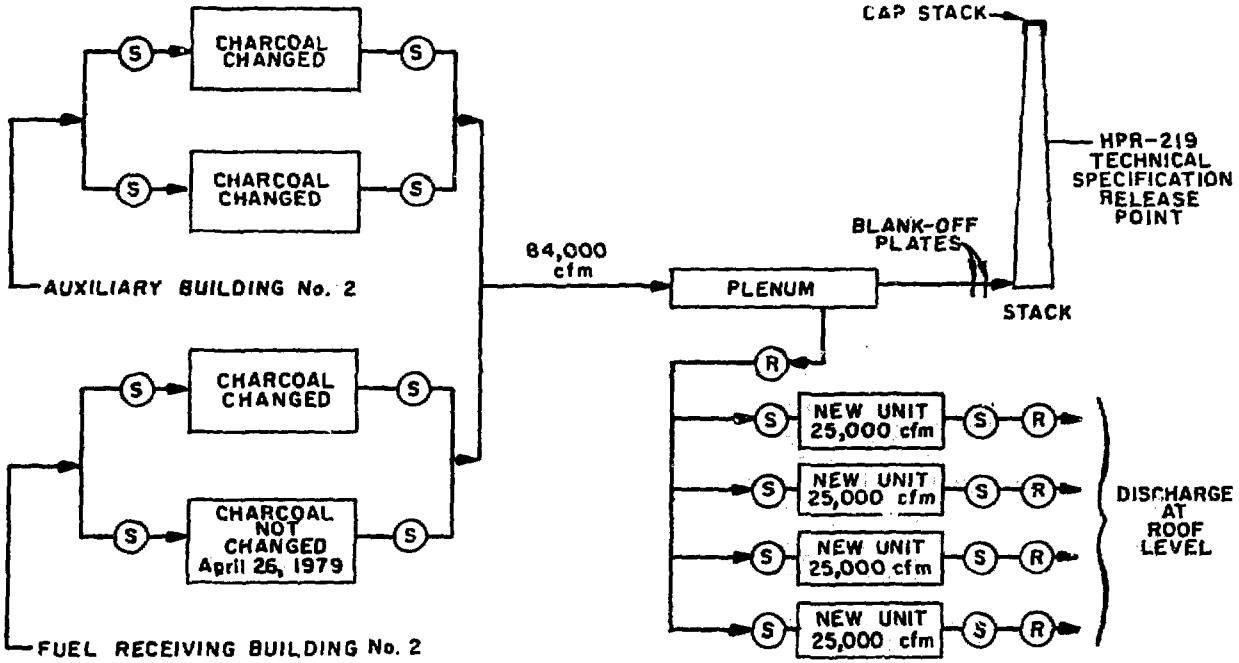

Fig 2. Off-gas system modifications.

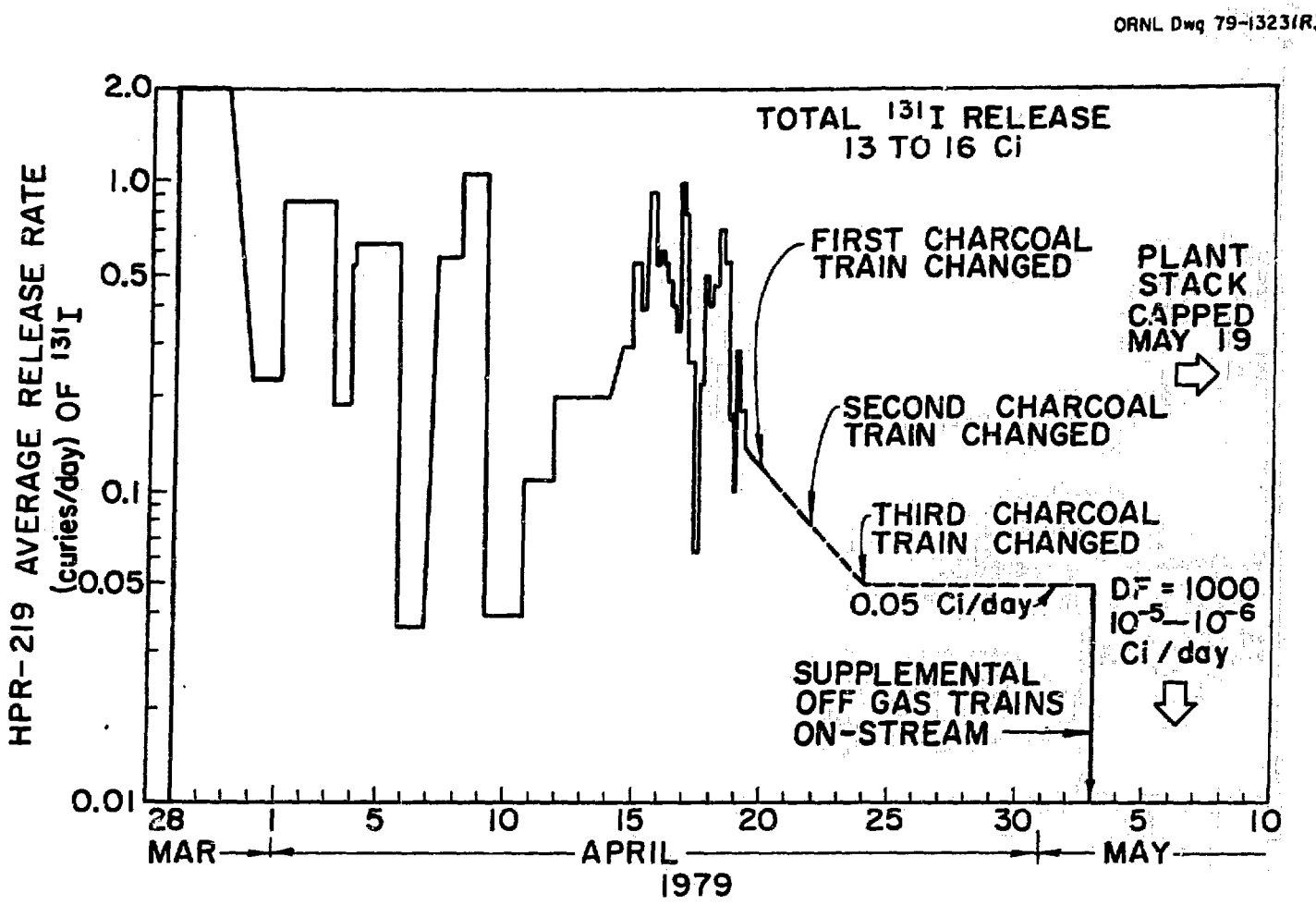

Fig. 3. Release of ^{131}I.

As of April 1980, a total of 44,000 to 53,000 Ci of ^{85}Kr are present in the Containment Building which must be removed to permit cleanup of the reactor to proceed. Serious consideration of this problem has been given to the various technologies which may be applicable, including (1) purging the reactor building; (2) charcoal processing; and (5) selective absorption. In a recently issued NRC environmental assessment,[3] it was concluded that purging the ^{85}Kr from the system is the most expeditious method for removal. It also results in the greatest environmental impact in terms of public dosage during normal operations, even though such doses are well within the established regulations. The other technical alternatives, recognizable chemical engineering unit operations, would take much longer to implement and would also require long-term storage of large quantities of charcoal containing ^{85}Kr or long-term sotrage of pressurized gas in piping and vessels. Inherent in these storage methods is the risk of subsequent accidental releases of krypton due to the failure of the storage containers or operator error.

From a technological standpoint, the selective absorption process represents an interesting challenge to chemical engineers, and proposals have been made to DOE to use this process as a mobile radwaste off-gas treatment system for emergency application. From a personal point of view, I believe that the development of this process (originally for the reprocessing of spent reactor fuels) through

the stage of theoretical unit operation and subsequent pilot-plant operation is an excellent example of chemical engineering. Dr. M. J. Stephenson of the Union Carbide Nuclear Enrichment Technology Division will present a paper later in this session outlining the applicability.

Control of Liquid Effluents

As is well known, large volume 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 role of the chemical engineer.

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 inoperative some vital instruments. 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. 4. The locations of pertinent areas are designated in Fig. 5. The primary reactor coolant loop contained 87,000 gal of highly radioactive coolant with an ^{131}I inventory of ~6,000,000 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.

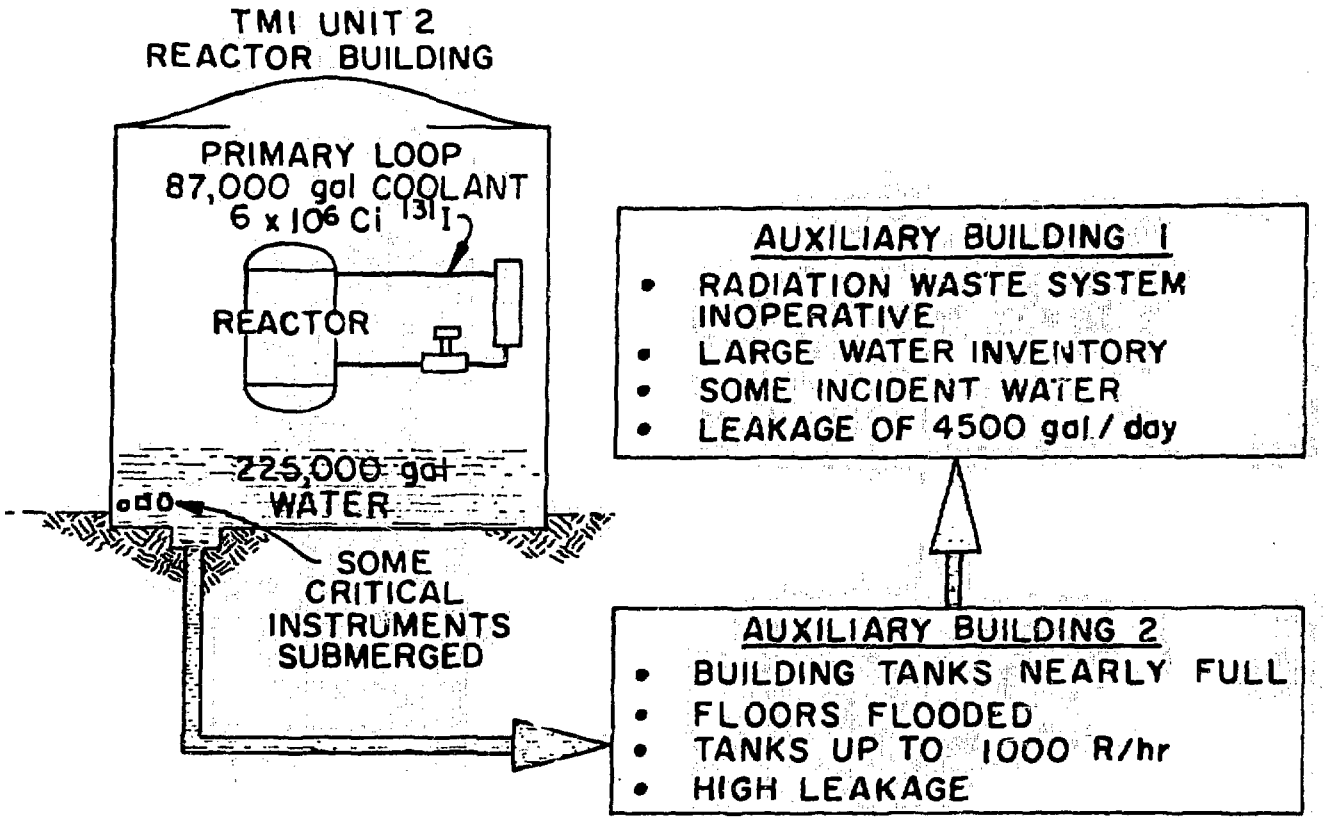

Fig. 4. Status of liquid at TMI following the accident.

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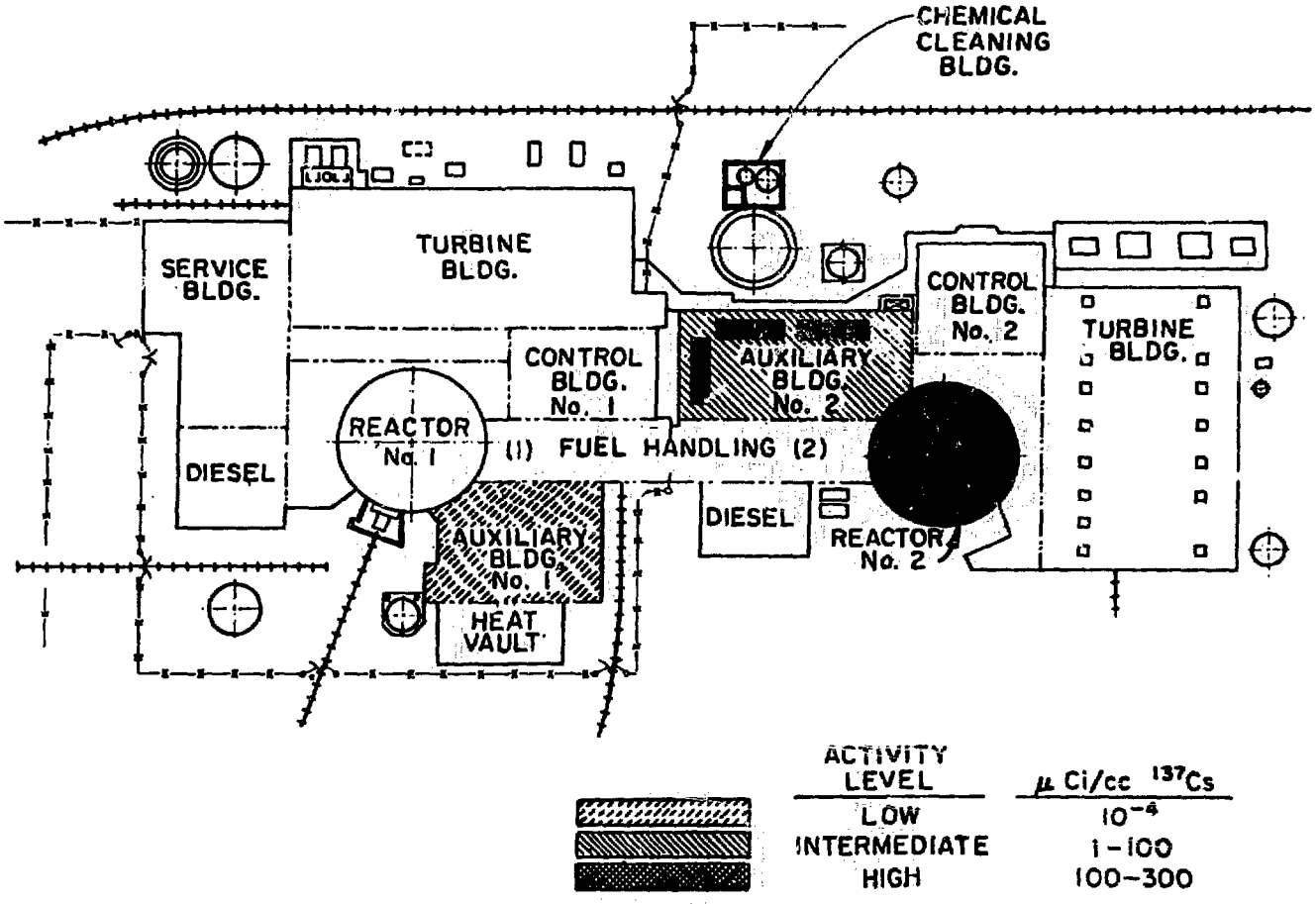

Fig. 5. Distribution of contaminated liquid

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 evaporator; 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.

Low-activity-level water

Low-activity-level water (LALW) was originally defined as all water from Unit 1 and any pre-event water, as confirmed by analysis, in Unit 2. However, when analysis of the Unit 1 water revealed that some Unit 2 post-event water had inadvertently been transferred into the Unit 1 Auxiliary Building vessels, the definition of LALW was modified to include any water that had an ^{131}I activity of <0.1 μCi/ml and contained no actinides.

A demineralizer system consisting of a filter followed by a mixer-bed demineralizer for activity removal was set up on the west side of the Unit 1 Fuel Handling Building to process Unit 1 LALW. This system was referred to as [illegible]r-I (initially Cap-Gun I) because it was being operated by Capolupo & Gundal, Inc., a subsidiary of Epicor, Inc. Two 20,000-gal Haliburton tanks were available for the decontaminated water. The first-pass decontaminated water went into one Haliburton tank and was sampled. If the water had not been decontaminated sufficiently to permit release after one treatment cycle, the filter and mixed demineralizer beds were changed and a second decontamination run was made. The second-pass decontaminated water was routed to the second 20,000-gal Haliburton tank.

Intermediate-activity-level water

Most of the tanks in the Unit 2 Auxiliary Building appeared to contain some post-accident water. This was verified by analysis of available samples of the tank solutions, which confirmed the presence of ^{131}I. Radiation readings generally ranged from low levels to a few hundred R/hr; however, the dose rates near the three reactor cooland bleed tanks exceeded 1000 R/hr. Thus, except for the water in the reactor coolant bleed tanks, all of the water in the Unit 2 Auxiliary Building tank was referred to as intermediate-activity-level water (IALW). The total volume of IALW ranged from 150,000 to 200,000 gal.

It appeared that most of this water could be treated sufficiently using a modified version of the Epicor system to permit discharge to the Susquehanna River. A containment system would be required to prevent the release of radioiodine to the atmosphere, and additional shielding would be required for handling the loaded demineralized beds. Furthermore, such a processing system could later serve as a final process step (i.e., a polishing step) for the high-activity-level water that would be subsequently treated.

A physical search of the Unit 2 facilities disclosed that the Chemical Cleaning Building could be used for housing a water treatment system. It not only met the space requirements but also contained two large tanks (one with a 90,000-gal capacity and the other with a 120,000-gal capacity) which could be used.

The system and equipment were collectively referred to as Epicor-II; the conceptual process flowsheet is shown in Fig. 6. The features that distinguished Epicor-II from Epicor-I were the containment provisions, the additional shielding of the loaded demineralizer beds, and the additional stage of demineralization provided by a second demineralizer bed in series. The Chemical Cleaning Building was sealed by spraying the interior surfaces with several layers of vinyl paint and securing some ventilation louvers. A 20-ton monorail crane was provided for remote handling of the loaded demineralizer beds; a remote control room was also included. All process equipment was vented to the building atmosphere through HEPA

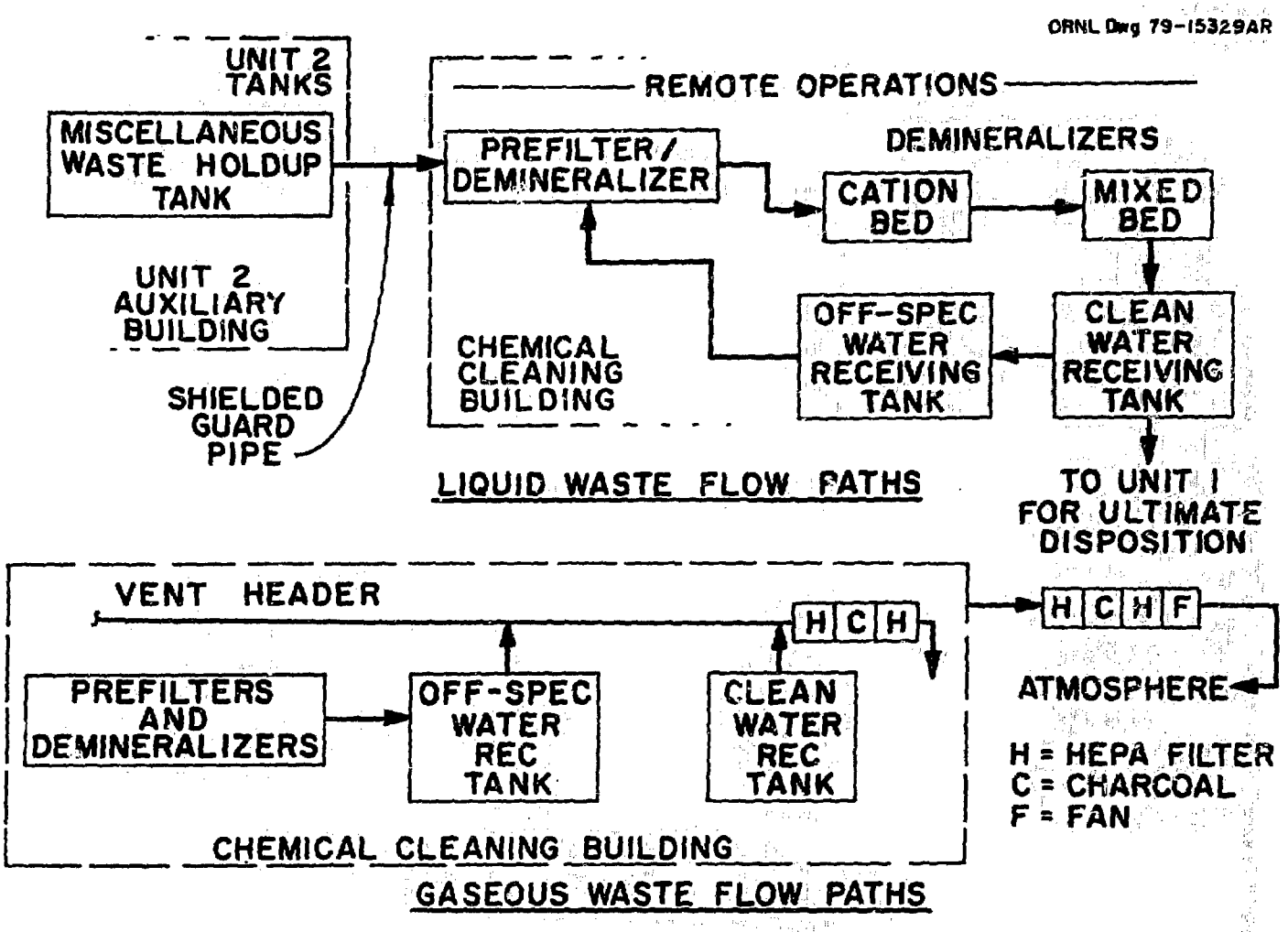

Fig. 6. Flow schematic of Epicor-II Processing system.

filters and a charcoal absorber, and the building was equipped with a 8000-cfm off-gas system.

High-activity-level water

The water in the Unit 2 primary reactor coolant loop and the Reactor Containment Building was collectively referred to as high-activity-level water (HALW). The total volume of HALW is in excess of 0.6×10^6 gal. Its outstanding characteristic with regard to treatment (or even transfer) was the very high ^{131}I concentration, which was estimated to be of the order of 10^4 μCi/ml. Radiochemical engineers recommended strongly that every effort be made to avoid disturbing any HALW until the radioiodine had been allowed to decay. Figure 7 shows the decay curves, based on an analysis of the primary coolant, for the radioactive nuclides that are of greatest concern in regard to treating that solution. It was realized, of course, that there could be overriding reasons for moving some solution, such as ensuring the continued operability of instruments that were vital to monitoring or controlling the status of the damaged reactor.

The problems of HALW processing were addressed early in the emergency by chemical engineers working with senior flowsheet and analytical chemists. A simplified schematic flowsheet is presented in Fig. 8 of the process under consideration for the treatment of HALW solution. This work has been continued throughout the past

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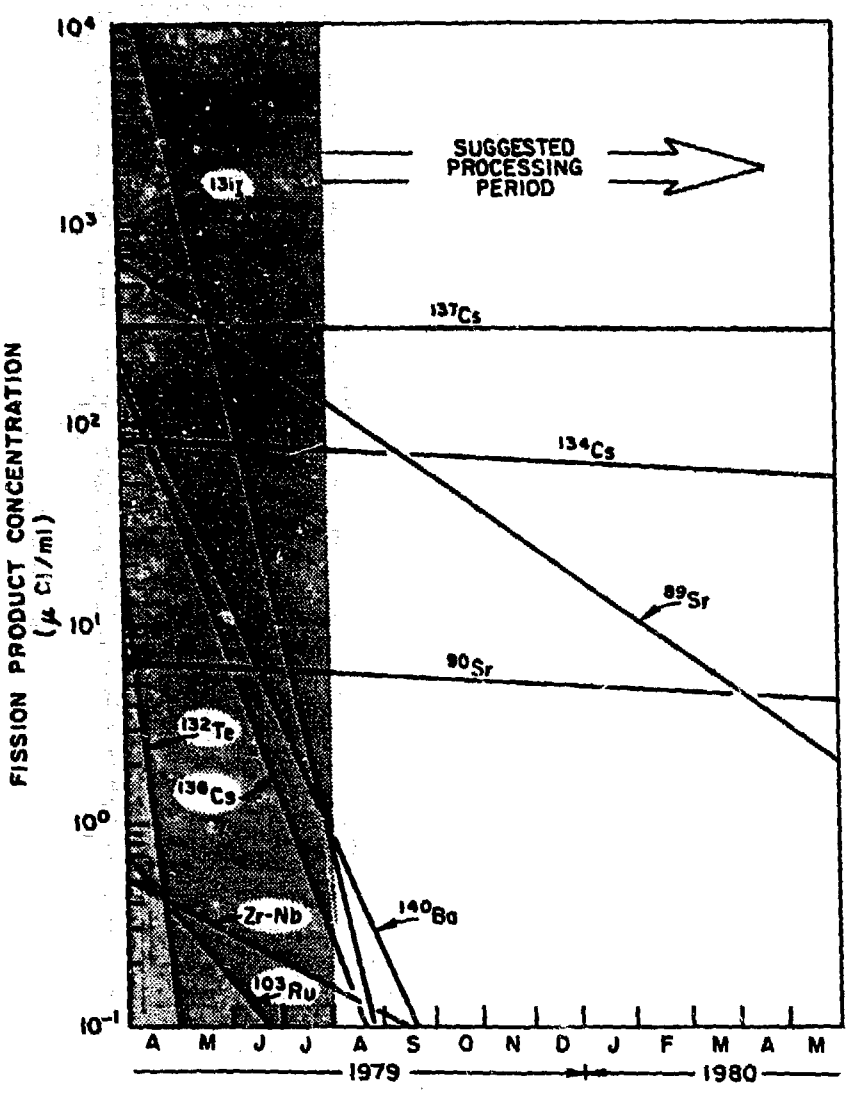

Fig. 7. Fission product decay in the TMI-2 primary loop.

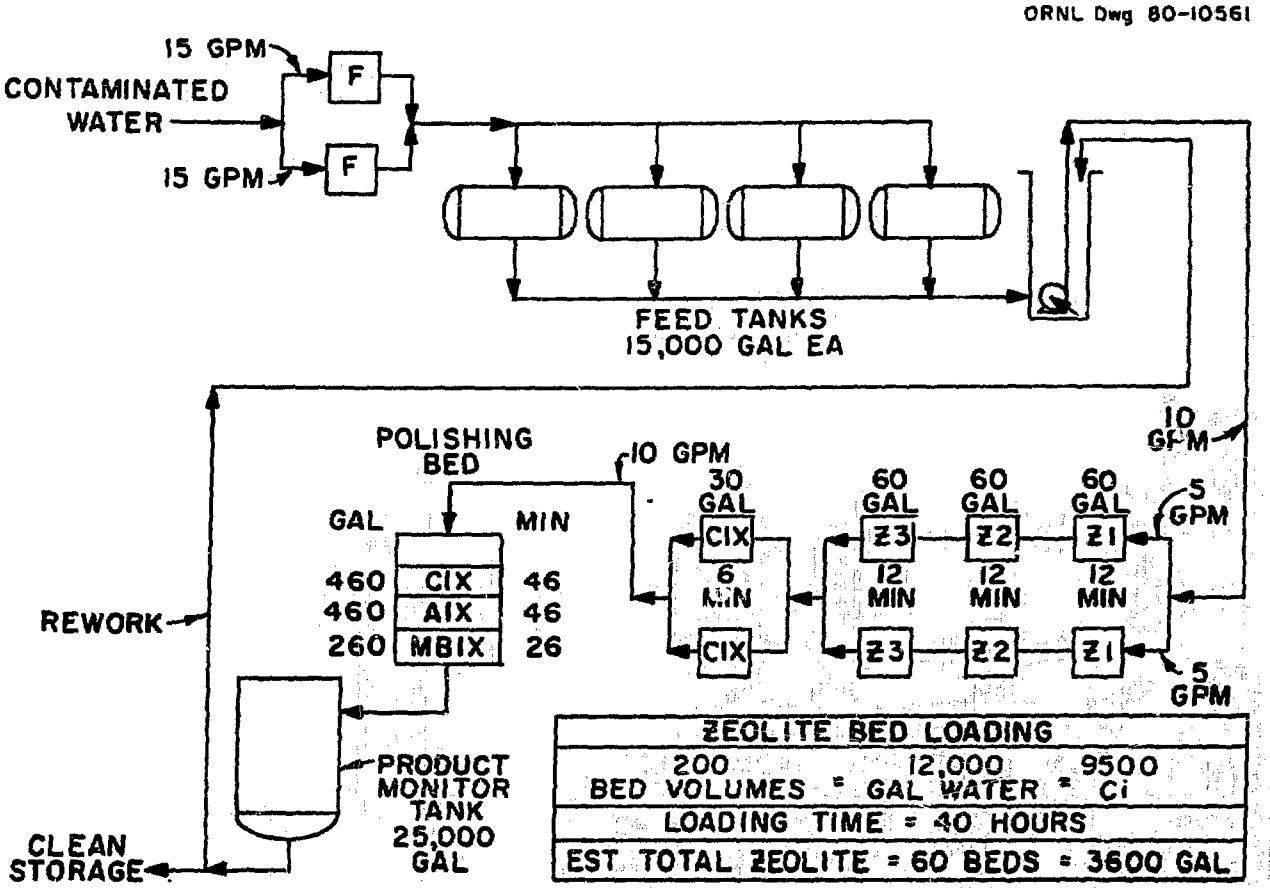

Fig. 8. SDS FLOWSHEET

year. Studies in this area will be reported in this symposium by E. D. Collins.[5] Basically, the work consisted of:

1. Providing conceptual engineering guidance for additional storage space of water from the Containment Building. There was considerable concern that the water level in the Containment Building might rise high enough to preclude operation of some instruments that were vital to monitoring and controlling the reactor.
2. Investigating and locating an evaporator to reduce the volume of contaminated water. It was presumed that the condensate would be sufficiently decontaminated to permit its discharge either directly or following subsequent treatment with the Epicor-I or Epicor-II system.
3. Providing assistance, including experimental work, in the development of ion exchange process flowsheets for the treatment of HALW.
4. Conducting an engineering study of an alternative process for HALW treatment.
5. Verifying the flowsheet finally selected and designed by Chem Nuclear Systems, Inc., and the Allied-General Nuclear Services using actual TMI water.

It is obvious from the foregoing discussion that the attention of chemical engineers will be required in the treatment of water resulting from an accident. An interesting proposal has been made to

DOE to provide a mobile liquid radwaste system capable of handling the large quantities of water that are generated in an emergency. One such proposal will be discussed by J. W. Snider[6] later on in this symposium.

Post-Accident Sampling

In addition to the control of effluents, a considerable amount of work is required to provide adequate sampling and analytical services to a damaged reactor. Work of this nature has been recognized by several utility companies in an effort to comply with a recent series of NRC requirements which were formulated as a result of the "lessons learned" at TMI. This subject will also be presented[7] in this symposium by G. E. German of the Tennessee Valley Authority (TVA) and W. J. Armento of ORNL.

Conclusions

From the foregoing background on the experiences gained from the on-site experience at TMI, it is apparent that the role of chemical engineers should increase in order for the nuclear option to proceed in a safe and efficient fashion. It is also obvious that as the results of the reports investigating the causes and effects of the accident come to light and attempts to backfit system designs to prevent a recurrence are studied, more technical demands will be placed on the profession.

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