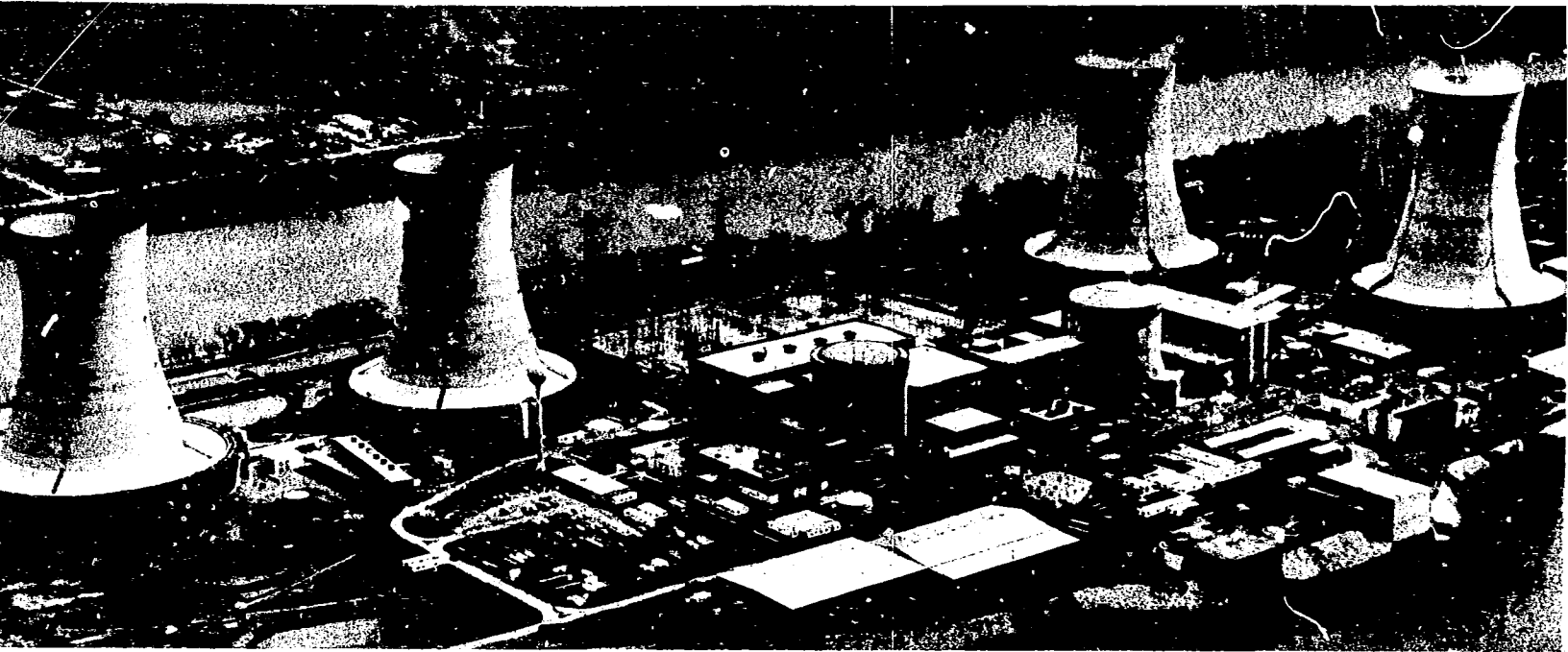This is an informal report intended for use as a preliminary or working document

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ESTIMATED SOURCE TERMS FOR

RADIONUCLIDES AND SUSPENDED PARTICULATES

DURING TMI-2 DEFUELING OPERATIONS; REPORT ON PHASE I

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ABSTRACT

The selection of methods for removal of the plenum and fuel from the Three Mile Island Unit 2 (TMI-2) reactor vessel will necessarily consider the quantities of radioactivity in solution and associated with suspended particulates in the reactor coolant. The total amount of suspended solids, which affects water clarity and equipment wear, must also be considered in the selection of the defueling technique and the subsystems needed to maintain acceptable operating conditions. Currently available data on relevant parameters are reviewed as the first phase of a project to develop estimates of source terms for soluble radionuclides, particulate radionuclides, and total suspended particulates during TMI-2 defueling operations. The approaches to be used in developing these estimates during Phase II are discussed in general terms. Additional measurements that could improve the quality of the estimates are recommended.

FOREWORD

This report is an account of work performed by Science Applications, Inc. between August 25 and September 30, 1981 on Phase I of Task Order No. 29, Subcontract No. K-1810 for EG&G Idaho, Inc. The ultimate goal of the project is to develop engineering estimates of the source terms for radionuclides which will contribute to personnel exposure and for suspended particulates during TMI-2 defueling operations. Work on Phase I concentrated on identification of potentially useful data and calculational techniques and development of rational frameworks for making the desired source term estimates. In Phase II, the available information will be analyzed in detail, the source term estimates and the associated uncertainty bounds will be developed, and the effectiveness of some potential control measures will be estimated.

ACKNOWLEDGEMENTS

CONTENTS

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DURING TMI-2 DEFUELING OPERATIONS
REPORT ON PHASE I

INTRODUCTION

The radionuclide and suspended particulate source terms encountered will affect the Three Mile Island Unit Two (TMI-2) defueling operations in at least three ways. First, the levels of suspended and dissolved radionuclides will affect radiation dose rates and airborne radionuclide concentrations. These in turn will dictate worker stay times and respiratory protection requirements. Secondly, suspended particulates may impair visibility and cause delays until conditions improve naturally, or until turbidity control systems provide adequate removal. Thirdly, suspended particles, if abrasive, may bring about the gradual deterioration of moving parts in mechanical devices used for defueling and may cause failures of elastomeric seals. Water clarity, dose rates, and respiratory protection requirements are all related to worker productivity on the job. These factors thus may strongly influence selection of the method of defueling, the design of equipment, the number of workers needed, the total radiation exposure during defueling, and the cost of defueling.

The ultimate goal of this work is to develop engineering estimates of the source terms for soluble and suspended radionuclides and for visibility impairment by suspended solids during TMI-2 defueling operations. The estimates developed will assist those responsible for defueling operations in the selection of acceptable defueling methods and specific subsystems needed to complete the work in an optimal manner. Work on the first phase of the project, reported here, identified sources of potentially useful data and calculational techniques and developed rational frameworks for making the desired source term estimates.

The next section identifies sources of information that are currently available and briefly discusses their quality and potential utility. Information expected from work that has been planned or is now in progress is identified. Additional measurements that would be useful are recommended.

These measurements are believed to be feasible in the short term (4-5 weeks) and could improve the estimates to be made in Phase II. Finally, the approach to be used in Phase II is outlined. The recommended methods reflect the amount and quality of information likely to be available during Phase II, scheduled for completion by November 30, 1981.

CURRENTLY AVAILABLE DATA

Information sources that have been identified are summarized below by topic. The sequence of topics begins with the radionuclide inventory in the core at shutdown, proceeds through processes affecting the fission product and particulate source terms, and ends with potential control techniques.

Radionuclide Inventory

The inventory of radionuclides in the TMI-2 core at shutdown is ultimately the source of the radionuclides that will be encountered during defueling. Several calculations of the inventory have been performed.[1,2] Reference 1 contains a comparison of the outputs of (a) the latest version of the ORIGEN computer code (Version 2), and (b) the LOR-2 code, a modification by Babcock and Wilcox of the ORIGEN code. For the principal nuclides of interest at the time of defueling, the shutdown inventories predicted by the two codes agree to within about 6%. The results of calculations using the CINDER-10 and EPRI-CINDER codes to estimate decay heat generation in the TMI-2 core are given in Reference 2. That report also contains some information on the core inventory by isotope. Because the tabulation emphasizes decay heat producing isotopes, it is less complete than the LOR-2 and ORIGEN-2 tabulations in Reference 1. The available CINDER results are also in good agreement with those from the ORIGEN code.

In Table 1, the shutdown inventories are given for 17 isotopes of interest. The tabled values are those computed by ORIGEN-2 except for ^{241}Pu and ^{241}Am, for which the LOR-2 calculations are known to be better.[1] The differences between the ORIGEN-2 and LOR-2 estimates for the other nuclides will not significantly affect the conclusions of this study. Most

TABLE 1. PARTIAL LISTING OF CALCULATED RADIONUCLIDE ACTIVITIES FOR TMI-2 AT SHUTDOWN AND AFTER DECAY

Radionuclide	Half-Life[b] (yr)	Radionuclide Activity (Ci)[a]	
		At Shutdown ($t_d = 0$)	After Decay ($t_d = 63$ months)
^{3}H [c]	12.33	4.1×10^3	3.1×10^3
^{85}Kr	10.7	9.7×10^4	6.9×10^4
^{90}Sr-Y	28.8	7.5×10^5	6.6×10^5
^{106}Ru-Rh	1.01	3.3×10^6	9.0×10^4
^{125}Sb	2.7	1.2×10^5	3.3×10^4
^{134}Cs	2.062	1.6×10^5	2.7×10^4
^{137}Cs	30.17	8.4×10^5	7.5×10^5
^{144}Ce-Pr	0.778	2.5×10^7	2.3×10^5
^{147}Pm	2.6234	2.6×10^6	8.1×10^5
^{151}Sm	90	1.1×10^4	1.1×10^4
^{155}Eu	4.9	3.2×10^4	1.5×10^4
^{238}U	4.468×10^9	2.7×10^1	2.7×10^1
^{238}Pu	87.74	7.3×10^2	7.6×10^2
^{239}Pu	2.41×10^4	8.6×10^3	9.0×10^3
^{240}Pu	6.57×10^3	2.4×10^3	2.4×10^3
^{241}Pu	14.4	2.0×10^5	1.6×10^5
^{241}Am	433	2.1×10^1	1.9×10^3

a. The quantity of t_d is the decay time.

b. Half-lives were taken from and are given with the same number of significant figures as in Reference 3.

c. An additional 200 Ci is estimated to have been produced by neutron activation reactions in the coolant during power operation.

of the isotopes listed have one or more of the following properties: large activity after 63 months of radioactive decay, significant γ-emitter, significant β-emitter, significant α-emitter, and potential airborne radioactivity hazard. Tritium (^{3}H) is listed because of its importance to our understanding of the behavior of other nuclides. Its presence as tritiated water provides a nonreactive tracer of fluid movement. Preliminary schedules show the start of defueling in mid-1984, approximately 63 months after shutdown. The inventories listed for that decay time reflect only the changes due to decay of the radionuclides listed and their precursors. The decayed inventories were hand-calculated using the ORIGEN-2 output[1] and the half-lives[3] shown in Table 1. If defueling is delayed, the activities of all nuclides except ^{241}Am will be smaller than the tabulated values.

Post-Accident Condition of the Core

Croucher has summarized the various lines of evidence used to estimate the post-accident status of the TMI-2 reactor core and the conclusions drawn from the available information.[4] There is general agreement that the core damage was substantial. Virtually all the fuel rods suffered cladding failure. It is estimated that 40-60% of the cladding was oxidized. Although it is believed the fuel did not melt, other components did. It is believed that a bed of fused core debris lies on top of the structurally intact stubs of fuel rods and that a second bed of loose fragmented core material lies on the fused debris. The particle size of the core debris can only be estimated. Fuel particle size distributions from a variety of experiments are plotted in Reference 4. Median particle size distributions range from 0.1 to 3 mm, with most of the values near 0.2 mm. Estimates of the total mass of loose and fused core debris range from 32000 to 58000 kg. The debris had caused 60-90% blockage of the normal upward water flow through the core.

The massive fuel damage described in Reference 4 would be accompanied by substantial releases of volatile radionuclides. Large fractions of the noble gases, radioiodines, and radiocesiums are known to have escaped from the primary system into the reactor containment building. Measured concentrations of radionuclides in various fluid samples have been used to

4

estimate the fission product releases from the fuel. The estimated release fractions for volatile radionuclides tabulated in Reference 4 generally confirm damage estimates based on thermal-hydraulic and hydrogen generation calculations.

Post-Accident Distribution of Radionuclides

The post-accident distribution of radionuclides is important for estimates of the defueling source terms. Releases of radionuclides from the fuel and from the primary system during and since the accident have reduced the potential source terms during defueling operations. These effects are particularly important for the volatile nuclides. Data from a variety of sources[5--16] are available to estimate the net activities in the fuel and in the reactor coolant system (RCS).

The RCS has been sampled approximately once a week since the time of the accident. Routine analytical results for ^3H, ^{85}Kr, ^{89}Sr, ^{90}Sr, ^{134}Cs, and ^{137}Cs are available.[5] In addition, data on short-lived noble gases, radioiodines, ^{136}Cs and ^{140}Ba were reported when these radionuclides were present in significant quantities.[5] Concentrations of chemical constituents of the RCS that have been determined routinely are B, Na^+, Cl^-, H^+, O_2, H_2, and N_2. A few of the RCS samples have been split and analyzed by several laboratories to determine uranium and plutonium isotopic concentrations and the concentrations of other radionuclides.[6,7]

Most of the RCS samples are taken from the cold leg of the RCS between the outlet of steam generator A and the inlet to reactor coolant pump-1A (RCP-1A). Approximately 30 gallons of liquid are purged through the line prior to sampling, which is adequate to clear the sampling line and the letdown line. However, it is not certain that the samples obtained are representative of the entire RCS. There is no forced coolant flow through the core, and the decay heat power level has declined to ~0.04 MW, simultaneously reducing the amount of convective flow. The RCS makeup flow is injected on the outlet side of RCP-2A. Whether the limited convection, diffusion, and the leakage-makeup flows provide adequate mixing is an open question. The data are only suggestive: (a) results from the few samples of the RCS taken from the pressurizer agree with those for the normal sampling location, and

(b) the change of ^{3}H concentration in the RCS is consistent with the rate of leakage. Both facts suggest that the samples are representative.

Samples of water in three reactor coolant bleed tanks (RCBTs) and of the large volume of water in the reactor building basement have also been obtained and analyzed in detail.[6,8] These fluids contain most of the radionuclides in liquids that have escaped across the primary system pressure boundary. Therefore, the measured radionuclide concentrations provide a basis for estimating the radionuclide activity still contained in the fuel and the RCS. The loss of reactor coolant through the relief valve during the accident and the RCS leakage rates since the accident have been estimated. That information, together with measured concentrations of radionuclides in the RCS, provides an alternative means for computing amounts of radionuclides outside the primary system.

Radionuclide Leaching

Radionuclides presently in the fuel debris will be leached into the RCS prior to and during defueling operations. The amounts of radioactivity leached will differ for each element. The leach rates for a number of radionuclides have been measured experimentally under laboratory conditions using samples of irradiated fuel.[17--20] The experimental leach rates are qualitatively similar and lead to a general leachability ranking of Sb, Cs > Sr > Ru, Ce, Eu, U > Pu. However, quantitative comparisons show the importance of particle size and leach solution on the results. Solutions that have been used are deionized water, distilled water, Hanford groundwater, synthesized brine, and simulated pressurized water reactor (PWR) coolant.

The simulated PWR coolant should in principle be most relevant to leaching from the TMI-2 fuel. It contained ~3300 ppm boron and ~1100 ppm sodium at a pH of 8.[17] Measured values of the same parameters in the RCS at TMI-2 have averaged 3760 ppm boron, 1070 ppm sodium, and pH 7.8.[5] The oxygen content of the solution, a possibly significant variable, was not controlled. At TMI-2, oxygen concentrations have been maintained at very low levels by control of water chemistry. The solution temperatures studied, 85 and 100°C, generally exceed the post-accident TMI-2 RCS temperatures which have ranged from 81°C in May 1979 to 46°C in June 1981. Temperature did not

appear to be an important variable in the 85-100°C range.[17] Leaching rates and cumulative release fractions (amount leached/original inventory) were measured for a period of 7.5 months. Ranges of release fractions were extrapolated for leaching times of up to 48 months. Further extrapolation, to 63 months, is necessary to estimate the total fraction leached prior to defueling of TMI-2. Ranges of estimated leaching fractions based on Reference 17 are given in Table 2. The ranges reflect differences in leaching for different particle sizes. Two principal fuel particle size ranges, between 17 and 18 mesh and between 100 and 200 mesh, were used in the experiments. The corresponding ranges of mesh openings are 1.4--1.0 mm and 0.15--0.074 mm, respectively.[21]

TABLE 2. CUMULATIVE LEACH FRACTION ESTIMATES
BASED ON REFERENCE 17

Element	Estimated Range for Fraction Leached in 63-Month Period
Sb	0.004 -- 0.02
Cs	0.003 -- 0.006
Sr	0.0008 -- 0.002
Ru	0.0002 -- 0.001
Ce	0.0002 -- 0.001
Eu	0.0002 -- 0.001
U	0.0001 -- 0.002
(Pu)[a]	0.00005 -- 0.004

a. Gross alpha radioactivity was measured and assumed to be representative of Pu.

A second source of information on radionuclide leaching prior to defueling is the existing set of measured concentrations of radionuclides in the RCS at TMI-2. As indicated in the previous section, long term sequential data have only been reported for radiocesium and radiostrontium isotopes. The ^{137}Cs concentration data were examined for two time periods, 120-481 days and 481-810 days after the accident. In both periods the concentration can be described by a function

$$C(t) = C(0)e^{-\lambda t} \tag{1}$$

where

$C(t)$ = concentration (μCi/ml) at time t

t = time (days) after the start of the interval

$C(0)$ = concentration at the start of the interval ($t=0$)

λ = effective removal rate constant (day^{-1}).

A least squares regression curve was fit to each set of data to determine the effective removal rate constant λ. The best-fit removal rate constants for ^{137}Cs during the two periods were 0.0024 day^{-1} and 0.0015 day^{-1}, respectively. The ^{90}Sr concentration has behaved very differently; it remained approximately constant for the entire period.

The implications of these measurements are clearer after examining the measured ^{3}H concentration. The ^{3}H present as tritiated water is a nonreactive tracer for the RCS. The tritium concentration pattern can also be described by Equation (1) and the removal rate constant can be interpreted as the RCS leak rate constant. The best-fit rate constants for ^{3}H in the RCS were 0.0030 day^{-1} for 120-481 days, and 0.0016 day^{-1} for 625-810 days. Reported ^{3}H concentrations in samples taken during the period 481-625 days varied excessively and were not used in the best-fit evaluation. The ^{3}H data analysis suggests that the RCS leak rates for the two periods were $\sim$260 gallons/day and $\sim$130 gallons/day, respectively. Although the ^{3}H data are variable, the use of two different leak rates appears justifiable and is more realistic than the previously reported constant leak rate of 150 gallons/day for the entire period.

The fact that the removal rate constant for ^{137}Cs during the first period is lower than that for ^{3}H suggests that ^{137}Cs may have been entering the RCS

8

during that period. However, the two rate constants are probably not significantly different because of the variability of ^{3}H concentration data. Most of the Cs was released from the damaged fuel during the accident, and there is evidence for Cs leaching during portions of the period 0-120 days.[22] Therefore, even though the leach rates estimated for Cs in Reference 17 are relatively high, it is not surprising that no significant additions of ^{137}Cs to the RCS were observed.

In contrast, the constancy of the ^{90}Sr concentration implies a source of ^{90}Sr to replace that lost by RCS leakage. Average inputs to the RCS of 22 Ci/day during the first period and 11 Ci/day during the second period would be required to maintain the ^{90}Sr concentration. If the source of ^{90}Sr is the fuel, the implied cumulative ^{90}Sr leach fraction for the entire 690-day period is 0.016. This interim estimate of the cumulative leach fraction is about ten times the value estimated in Reference 17 for the first two years of exposure. Because the leach rate was probably higher during the period of 0-120 days, the final estimate of the cumulative leach fraction for the entire 810-day period will most likely be greater than 0.016, and the difference between this value and the estimate in Reference 17 will probably increase.

The magnitude of the difference between the two release fractions for ^{90}Sr calls into question the applicability of laboratory leach rate measurements to the TMI-2 situation. As suggested above, leaching of ^{90}Sr from the fuel may not be the source of the activity entering the RCS. If another process, such as redissolution of material previously released from the fuel, is controlling, then this comparison of the laboratory leaching data for ^{90}Sr to TMI-2 observations is not relevant. Comparisons of concentration changes for other radionuclides in the RCS and for all radionuclides in isolated tanks and lines containing coolant and fuel debris will help resolve whether the laboratory leaching data are representative for post-accident conditions.

The leach rates that have been observed are probably lower than those that will prevail during defueling because the exposed surface area and turbulence will be increased when the debris bed is being removed.

9

Suspended Particulates

Samples of the RCS have been described as "yellow" and "turbid" for more than a year. However, no measurements have been made of the optical properties of the samples. These general observations of turbidity are consistent with observations of suspended particulates in stored borated water in operating PWRs. The contents of refueling water storage tanks have been found to be turbid, even though the water was thoroughly filtered after a previous use.[23]

The size distributions of suspended particulates have been measured for two samples of the RCS. The data suggest that between March 29, 1979 (Sample RCS-1) and August 14, 1980 (Sample RCS-2) a significant amount of settling of larger particles occurred. The median particle size changed from greater than 3 μm to less than 1 μm during that period. During the same time interval, the total solids concentration increased by almost a factor of two.[6] The mechanisms responsible for these changes have not been identified.

The principal constituent identified[6] in the solid portion of samples from the RCS and the three reactor coolant bleed tanks (RCBTs) was iron, with Fe_3O_4 specifically identified by x-ray diffraction in RCS-1. The elements Ag, Cr, In, Na, Ni, Sn, and Zr were detected in RCS-1 solids but not in those from RCS-2, and the concentrations of Al and Ca were apparently lower in RCS-2. However, it must be noted that only 85% of the solids from RCS-2 (the 0.45-- 1.2-μm fraction) was analyzed, so the results may be slightly biased. Concentrations of fissionable nuclides in solids also decreased substantially during the period between the two samples. Settling of the larger particles is thought to have occurred, although the concentration of dissolved iron was much higher in RCS-2 than in RCS-1.

Telephone contacts were made with personnel in operating power plants to obtain information on operational problems encountered during routine refueling and remedial actions taken to overcome the difficulties. The information obtained was qualitatively similar to that documented in a 1979 study conducted for the Electric Power Research Institute.[23] The principal sources of turbidity were particulate material released from primary system surfaces and particulates in the water from borated water storage tanks. Releases of particulates from primary system surfaces were related to oxygen concentration,

pH and temperature of the RCS, and to the flow path of the refueling water. Some measurements of corrosion rates and corrosion product release rates under a variety of conditions have also been made.[24] These will aid estimates of the potential release of particulates that could impair visibility. Releases of corrosion products were found to be increased by flooding the cavity through the core (i.e., via the cold leg of the RCS) following shutdown.[23]

Debris generated during underwater thermal, abrasive, or mechanical cutting operations that may be required to remove the plenum assembly is another source of particulates in the cavity water that could potentially interfere with vision during defueling. Information on the amounts and size distributions of particles generated by these operations is being sought.

It is clear from (a) the expected condition of the TMI-2 core, (b) descriptions and analyses of the RCS samples, (c) the operational experience during routine refueling described in Reference 23, and (d) nonroutine operational experiences with other fuel pools[25,26] that turbidity from defueling operations will be a problem that will require careful control. The movement and breakup of core debris will certainly increase the levels of suspended particulates. Secondary problems associated with particulates include (a) deposition on cavity walls followed by increased radiation exposure rates and/or resuspension of radionuclides into the reactor building air, (b) the formation of surface flocculant that also impair visibility, and (c) greater mechanical wear and degradation of components to be operated underwater. It may be possible to conduct representative short-term tests to evaluate the effects of wear on metallic and elastomeric components using a synthesized suspension of particles. Additional data are needed to identify the type(s) of particles most likely to affect such systems.

Control Techniques

The factors discussed above require that systems for controlling turbidity be available to maintain adequate water clarity during defueling. Because the duration of any turbidity problem depends on the ratio of cleanup flow rate to the total volume being treated, careful consideration must be given to the system flow rate and a balance struck between system size and cost and the

desired cleanup time. As noted in the previous section, the direction of fluid flow is also an important consideration.

In general, the original design of filtration and ion-exchange systems have been frequently found to be inadequate by power reactor plant operators. The inadequacies resulted from a lack of cleanup capability under certain conditions and from systems whose flow rates were too small for the volume of water that required treatment. Solutions to problems were evolved over a period of time at each plant. Submersible ion-exchange and filtration systems have been developed to provide localized cleanup capabilities to supplement the initial design capacities. Skimmer systems and stand-alone submersible filtration and ion-exchange systems have both proved useful under certain circumstances. Chemical treatment to reduce surface flocculants has been partially effective.[23]

The operation of these cleanup systems can be limited by pressure drop due to the buildup of particulates or by radiation limits. In normal use, the buildup of radionuclides has been more limiting than the pressure drop criterion. The period of operation prior to filter change could be a limiting factor in the effectiveness of such systems.

Vacuum techniques used to dislodge and subsequently collect particulate debris during defueling may lead to increased levels of soluble radionuclides. The debris may be fragmented further when being dislodged. Secondly, the filtered material will be in contact with flowing liquid which may lead, at least initially, to increased leach rates. The magnitude of this problem will depend upon the cleanup system configuration and the point of discharge. If an ion exchange bed is included in the effluent side of the particle collection system, such problems may be minimized.

Filling the cavity from above the core, rather than through the cold leg, has been found to reduce the turbidity generated during flooding.[23] In addition, maintenance of downward flow during defueling should reduce the amount of debris in the upper layers of the water. Refueling experience has shown that water clarity is improved by maintaining flow from the fuel pool through the transfer canal and deep end of the cavity and then into the reactor vessel. This practice also reduces transport of contaminants into the fuel storage pool during transfer operations.

If conventional control systems appear inadequate, one certain way to avoid turbidity problems in the vessel and cavity water is to perform the defueling operations after draining the RCS. This approach would require a hot cell on top of the reactor vessel and the capability to move core debris out of the vessel through airlocks. A secondary containment around the hot cell and airlocks would probably also be needed, together with adequate ventilation and air filtration equipment. The feasibility of this approach has not been analyzed.

DATA EXPECTED FROM WORK IN PROGRESS

It is desirable to obtain more information on the fuel debris particle size distribution and the abrasive properties of those particles. Possible sources of data are the solids from the makeup filters and RCBTs in the Auxiliary Building. These solids would reflect the smaller portion of the distribution of fuel debris and provide a bound on the minimum fuel particle sizes likely to be encountered. The same samples could be used to perform experiments to determine leach rates under a variety of conditions. One sample from the MU-F-5B filter has been sent offsite for analysis. It is possible that at least preliminary information on debris particle sizes will be available within the next month.

RECOMMENDED ADDITIONAL DATA COLLECTION

The following recommendations for additional data collection are limited to relatively simple tasks that could reasonably be completed in ~4 weeks and would be useful for determining the source terms during defueling.

1. Evaluate gamma spectra obtained by Babcock and Wilcox to determine whether concentrations for other gamma emitters in the RCS (e.g., ^{106}Ru, ^{125}Sb, ^{144}Ce) can be deduced from the spectra. Data on the other nuclides will help to determine whether input of those elements to the RCS has occurred, and if so, the rate of input. The fact that leach rate estimates based on laboratory data appear invalid makes the need for more "field data" imperative. It is more than confirmatory information; it may

be the only source of reliable estimates of leaching. (The spectrometer calibration information and counting geometries are needed together with the spectra obtained. Spectra of a few samples should be examined first to assure the feasibility of extracting the desired data. High radiocesium concentrations may preclude determination of the other nuclide concentrations without a chemical separation.)

2. Determine by optical measurement the absorption coefficient of unfiltered and filtered RCS samples collected at different times. Light sources similar to those in the existing and proposed cavity lighting systems should be used. These measurements will provide information on light and image transmission through the RCS and about changes in the RCS turbidity.

3. Obtain by filtration particle size and solids concentration information for additional samples of the RCS to determine whether the particle size distribution has stabilized or fluctuates. The same samples employed for item 2 above should be used for these measurements and measurements to determine the abrasive nature of the particles.

4. Conduct leach rate studies using fuel debris that has been collected from the makeup filters and RCBT-A. Samples of this debris have already been collected and could be used to measure leaching of fission products from the debris by simulated PWR coolant under quiescent and agitated conditions.

APPROACH FOR PHASE II

The source term estimates to be made in Phase II of this work depend upon further analysis of existing information and upon the collection and analysis of additional information as outlined in the previous section. The approaches to be followed are discussed below.

For soluble radionuclides, the distribution of radioactivity can be described by a two-compartment model with removal and internal cleanup

14

capability. For partially soluble fission products, a third compartment is needed to account for the activity in fission product precipitates that are separate from the fuel debris. Figure 1 is a diagram of the more complex model.
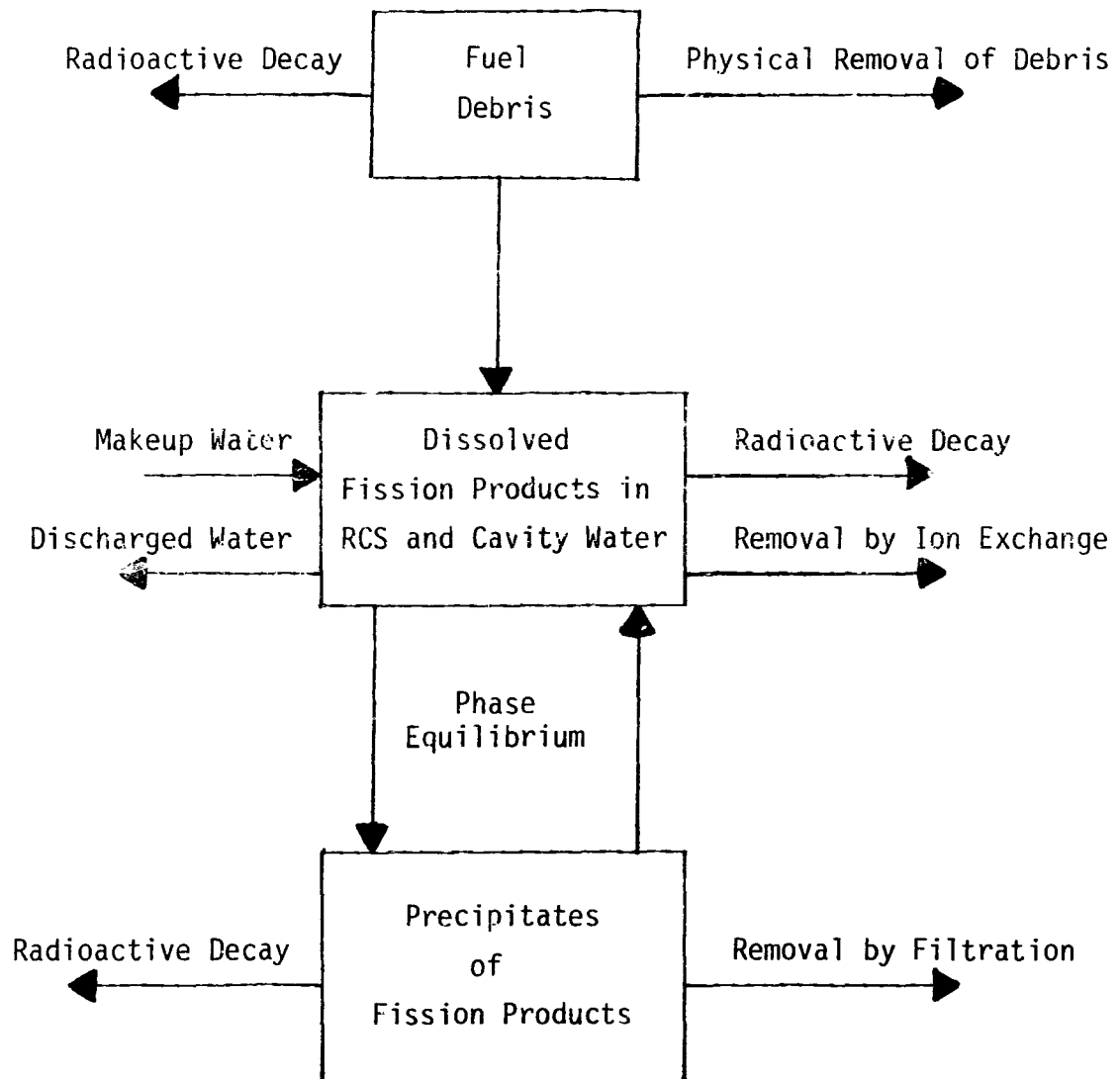

Figure 1. Conceptual Three-Compartment Model for Partially Soluble Fission Products

The same model can be used for the various distinct phases of the accident and the recovery process through defueling. A "phase" is operationally defined by significant changes in one or more of the rate constants that describe the system. To estimate the source term during defueling, it is necessary to examine the behavior of fuel and fission products during earlier phases. The sequence is outlined below.

1. <u>Inventory of radionuclides in fuel debris.</u> This is reasonably well established for ^{134}Cs and ^{137}Cs; however, it is presently uncertain for other important nuclides, notably ^{90}Sr. Analysis of data from isolated volumes containing fuel debris should help resolve whether the ^{90}Sr is in the fuel debris or present as a precipitate in the RCS.

2. <u>Previous and current leach rates.</u> Determination of the leach rates is of course closely related to Item 1. Evaluation of data from periods of apparently rapid leaching soon after the accident will aid estimates of leaching during defueling when substantial agitation of debris and fluids will occur. It is important to determine whether published estimates of leaching rates from laboratory experiments can be used or whether (as the ^{90}Sr data now indicate) such estimates are quite misleading. Verification of previous estimates of the RCS leakage rates is needed and can be performed.

3. <u>Projected changes prior to defueling.</u> Changes in the fuel debris and RCS inventories will be computed based on the estimated leaching rates and projected additions of uncontaminated water to the RCS. The results of the calculation will be the RCS and fuel debris inventories at the time defueling begins.

4. <u>Releases during defueling.</u> The inventories and estimated leach and removal rates during defueling will be used to estimate reactor cavity concentrations during defueling. These estimates will consider the effectiveness of cleanup systems and the potentially accelerated leaching of soluble species from debris in the particle collection and filtration systems.

For radioactive particulates, the approach will be similar. Steps 1.--3. above will also yield the inventories of radionuclides in particulate form. Fracture and resuspension of particulates will be sources of suspended radionuclides during defueling operations. Cleanup systems and leaching will decrease the concentrations of radionuclides present in suspended particulates.

For nonradioactive particulates, the data base for estimating concentrations is substantially less complete than that for specific radionuclides. A model similar to that shown in Figure 1 can be conceived, but the data base is probably not adequate to justify its use. As noted previously, additional measurements are needed to improve the estimates of turbidity during defueling. Data on the generation and release of corrosion products during normal operations are given in Reference 24. Additional data for crud generation under the extremely high temperature conditions during the accident will be sought to bound the mass of particulates available for resuspension during defueling. The effectiveness of potential particulate cleanup systems for maintaining adequate visibility will be estimated.

The uncertainties associated with the source terms will also be estimated during Phase II. It is expected that the possible range of source terms may be substantial. This is particularly true for turbidity because of the lack of measurements. Completion of the measurements recommended in the previous section will reduce these uncertainties.

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